

EGU21-10936

<https://doi.org/10.5194/egusphere-egu21-10936>

EGU General Assembly 2021

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The effect of melt water content and isothermal annealing time on the formation and evolution of clinopyroxene-titanomagnetite clusters

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Crystal clustering influences the formation of crystal mushes and the rheology and differentiation of magmas. Heterogeneous nucleation is known to be an important cluster-forming mechanism, but there has been little systematic experimental study of cluster formation and evolution.

In this study, we analysed dynamic crystallization experiments from Pontesilli et al. (2019), focusing on clusters of clinopyroxene (cpx) and titanomagnetite (tmt). These experiments aimed to reproduce the crystallisation behaviour of dry (nominally 0 wt.% H₂O) and hydrous (2 wt.% H₂O added) Etnean trachybasalt at mid-crustal storage conditions (400 MPa, 1100°C, NNO+1 oxygen buffer, corresponding to undercooling of 120°C and 80°C respectively). After superheating at 1300°C for 30 minutes, samples were cooled at 80°C/min to 1100°C and annealed for dwell times ranging from 0.5h to 8h.

Electron backscatter diffraction (EBSD) maps and image analysis were used to quantify clustering parameters such as tmt number density, “shared perimeter fraction” (“SPF”, the fraction of total tmt boundary length shared between cpx and tmt), “fraction of touching tmt” (“FTT”, the fraction of all tmt grains that are touching cpx), and the crystallographic orientation relationships (CORs) between cpx and tmt. Dry samples generally show a higher number density of tmt crystals than wet samples. SPF and FTT are highest (≥ 0.40 and ≥ 0.93 respectively) in the 0.5h duration dry experiments. Both parameters fall to ≤ 0.25 and ≤ 0.75 respectively after 4h of annealing. In wet experiments, SPF and FTT are lower (≤ 0.33 and ≤ 0.79 respectively) at 0.5h annealing time and do not decrease strongly with annealing.

EBSD maps reveal that $> 70\%$ of tmt grains are in contact with cpx in all analysed samples. Tmt exhibits two closely related CORs to cpx. More than 60% of total tmt-cpx boundary length in all samples follows COR 1 ($[-110]_{\text{tmt}}[010]_{\text{cpx}}$, $[111]_{\text{tmt}}(100)^*_{\text{cpx}}$, $[-1-12]_{\text{tmt}}[001]_{\text{cpx}}$) or COR 2 ($[-110]_{\text{tmt}}[010]_{\text{cpx}}$, $[-1-11]_{\text{tmt}}(-101)^*_{\text{cpx}}$, $[112]_{\text{tmt}}[101]_{\text{cpx}}$). COR frequencies suggest a strong influence of water content and annealing time on their formation. In the 0.5h duration dry experiment, tmt-cpx boundaries following COR 1 are twice as frequent by length as those following COR 2, whereas in the 0.5h duration wet experiment, COR 2 boundaries are 5 times more frequent by length than

COR 1 boundaries. In both wet and dry experiments the length ratio of COR 1 : COR 2 boundaries approaches 1 with longer annealing times.

The degree of undercooling (as imposed by the different water contents) is the most important influence on the microstructural clustering parameters, leading to lower overall number densities of tmt as well as affecting the SPF and FTT values at short durations and the subsequent evolution of these parameters with increasing annealing time. The high frequency of tmt-cpx CORs is consistent with heterogeneous nucleation. However, the mechanisms controlling which CORs develop are unclear. Annealing does not fully erase CORs or microstructural signatures of clustering, suggesting that crystal clusters erupted in volcanic products could still preserve signs of their formation.

Pontesilli et al. (2019), *Chem Geol* 510:113-129. 10.1016/j.chemgeo.2019.02.015

Funded by the Austrian Science Fund (FWF): P 33227-N