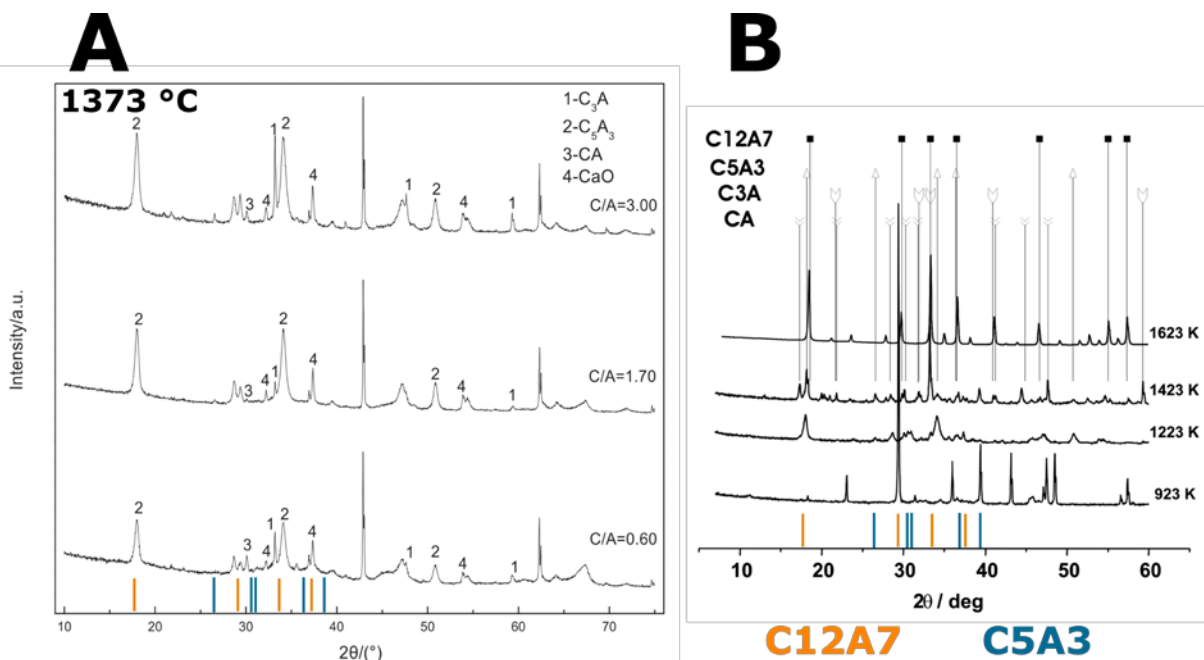


SF 1 Misidentification of C12A7 (orange) as C5A3 (blue) has led to the notion that C5A3 + C3A are the formation pathways to calcium aluminate compounds [2,3]. Depending on reactant heterogeneity C12A7 + C3A or CA + C3A are the formation pathways based on the referenced thermodynamic experimental studies with correct phase identification. In our kinetic study C3A + CA was the formation pathway with high and low reactant heterogeneity, and direct formation of C12A7 is the formation pathways with atomic homogeneity.



Supplementary Information Table 1 HTXRD
instrumental and data collection parameters

Instrument Parameters	
Instrument	PANalytical Empyrean
Source	Cu $K\alpha_1/K\alpha_2$
Stage	Anton Parr HTK 1200N
Detector	PIXcel3D-Medipix3 1x1 detector
Mode	Scanning Line Detector
Active channels	255 (3.3482 °2 θ)
Collimation	
Incident	
Divergence Slit [°]	1/4
Soller slit [rad]	0.04
Anti-scatter slit [°]	1/2
Mask [mm]	10
Diffacted	
Anti-scatter slit [mm]	5
Soller Slit [rad]	0.04
Scan Parameters	
Step Size [°2 θ]	0.0131
Step Time [s]	13.7
Scan Time [s]	90/150*
Scan Range [°2 θ]	27.5 - 37
Temperatures	Dynamically (RT to 1100/1200 °C)*

* PVA/SSS respectively