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Kinetics of carbon isotope fractionation and CaCO₃ crystallisation upon the setting of lime mortar

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An examination of historical lime mortars has shown that their calcium carbonate cement displays a rather wide range of carbon isotope compositions from $\delta^{13}C \approx -25$ to $-7 \, {}^{o}/_{oo}$ (PDB). Obviously, this range reflects various processes of isotopic fractionation which have occurred during the history of an individual cement. But it is not clear which of the data may represent a primary composition. In order to establish this composition experimental brickwork has been carried out. Surprisingly, the measurements show that calcite of lime mortar does not have a unique carbon isotope composition. The δ^{13} C values change systematically from -11 $^{o}/_{oo}$ at the outside of the brickwork to $-2 \circ /_{00}$ (PDB) within the interior. This zoning along a mortar layer is caused by a kinetic fractionation factor $\alpha_{CaCO3-CO2(a)} = 0.9960$ which is given by the absorption of atmospheric carbon dioxide into a strong alkaline slurry of portlandite, sand an water and its subsequent reaction to calcite within a liquid boundary layer surrounding the Ca(OH)₂ particles. Thus, the diffusion and the reaction of carbon dioxide leads to an enrichment of ¹³C over ¹²C at the gas-side of a reaction front so that the calcite becomes continuously "heavier" from the exterior to the interior along a mortar layer. The overall process may be described by a fractionation according to zone melting.