TITLE: A Novel Pattern of Reactive Crystallization of Calcium Carbonate Formed in Agarose Gel

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The formation or dissolution of $CaCO_3$ in nature, influenced by carbonate equilibrium, is impacted by rising atmospheric carbon dioxide levels. The Liesegang Phenomenon, a self-organization phenomenon, manifests as periodic patterns created by crystal bands within a gel medium. This study examines the effect of carbonic acid equilibrium on $CaCO_3$ formation by analyzing pattern formation during the reaction crystallization of Na_2CO_3 and $CaCl_2$. A glass test tube was filled with a mixture of Na_2CO_3 solution and agarose gel for solidification, and a $CaCl_2$ solution layer was deposited onto the mixture to crystallize $CaCO_3$ at 5–10 °C. The findings of this study revealed that the integration of high-frequency periodic patterns, comprising fine bands, formed low-frequency patterns—a novel Liesegang Phenomenon in calcium carbonate. Prior investigations into crystal formation within the gel indicated that the reaction between calcium ions and hydrogen carbonate ions led to the formation of low-frequency periodic patterns. Simulation using the Nucleation-Growth model with a high nucleation threshold produced a pattern similar to the previous experiment with $NaHCO_3$ and $CaCl_2$. This suggests that the reaction between calcium ions and hydrogen carbonate ions yields significant amounts of calcium hydrogen carbonate ions instead of $CaCO_3$, despite the use of Na_2CO_3. Moreover, it was proposed that calcium hydrogen carbonate ions influence the dissolved state of calcium carbonate, resulting in the coexistence of high- and low-frequency patterns in pattern formation. Therefore, investigating calcium hydrogen carbonate ions could shed light on whether $CaCO_3$ crystals grow or dissolve under elevated atmospheric carbon dioxide concentrations.