Crystal Formation on Collagenous Gelatins: Effects of Carboxylation and pH on Rates and Polymorphs of Calcium Carbonates

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Organisms synthesize skeletons by growing crystals within specialized tissues to produce structures that are optimized to meet functional needs. An understanding of how macromolecule chemistry controls crystal formation within collagens to produce complex composites may lead to crystal-modified synthetic tissues with medical and technological applications. The purpose of this study was to test two hypotheses to determine the effects of carboxylation and pH (independent variables) on the number and polymorphism of calcium carbonate (CaCO3) crystallites (dependent variables). Experiments used gelatin substrates, which are often considered model compounds for collagen. These were placed in flow-through chambers to monitor crystal development using CaCl2 and NaHCO3 solutions. The number and type of crystallites that formed were characterized with optical and scanning electron microscopy. At the highest pH of 9.90, calcite was the predominant polymorph in contrast to aragonite at lower pH. Independent samples t-test showed 1) substrates with higher carboxyl density promoted more calcite (p0.10) and more total crystals; 2) crystallization rate increased with pH for all substrates (p0.05). Aragonite was favored in environments with a combination of fewer carboxyls and lower pH. This finding is previously unreported. Further analysis uncovered a systematic relationship between rate of crystallite formation and carboxyl density. The findings demonstrate a chemical basis for regulating crystal formation in collagens to produce crystal modified tissues with enhanced material properties. This type of fundamental research may lead to future patent opportunities for synthesis of new bone substitutes and modified tissues for regeneration or controlled drug delivery.