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A multi-step model for the dissolution mechanism of calcium apatite in acids

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Introduction: A complete dissolution mechanism of calcium hydroxyapatite (HA) in acids is the key for the understanding of progression of mineral loss of enamel in caries. Various models examining differing aspects of both thermodynamic and kinetic aspects of HA dissolution in acids are found in the literature, but there is no definitive overall model. Such a model would be beneficial for understanding HA behaviour and design of active ingredients of prophylactic oral health care products.

Aim: The aim of this study was to analyse the available models of HA dissolution to identify the key aspects of each and combine these to produce an overall general dissolution model.

Methods: Eight different models of HA reacting with acids were critically reviewed. The identifying features were extracted from each model and used to generate a general multistep model incorporating the key features of each.

Results: HA dissolution appears to be a series of distinct identifiable steps of chemical kinetic and thermodynamic reaction processes, involving the reaction of acids with HA surfaces. These steps include (1) diffusion of acids from the bulk solution through the Nernst layer, (2) their adsorption onto the surface, (3) chemical transformations on the surface, (4) desorption of the reaction products, (5) their diffusion through the Nernst layer to the bulk solution. All the steps mentioned appear to be much more complex. For example, processes (1) and (5) include chemical transformations happen with the ions during diffusion, because solution pH is known to depend on the distance from the solid/liquid interface (in acidic solutions it is higher near the surface of apatite and decreases when the distance increases). Processes (2) and (4) include ionic diffusion along the surface both to (step 2a) and away from (step 4a) the crystal steps, respectively, as well as a diffusive jump (steps 2b and 4b). Finally, process (3) consists of several successive chemical transformations (step 3a) and includes ionic detachment from the kink sites (step 3b). Moreover, for adsorption (step 2) to take place, the following intermediate steps are necessary: (i) dehydration of a surface site, (ii) partial dehydration of ions and (iii) a diffusive jump toward the surface. One can also expect three similar intermediate steps (diffusion jump from the surface, hydration both the ions and the surface site) for desorption (step 4). It is clear that some of these steps require further experimental investigation.

Conclusions: A general description of HA dissolution requires a multi-step model of the individual diffusion and reaction steps.

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