

## Solubility and toxicity of hydroxylapatite (hap) nanoparticles (nps): Implications for nanobiomaterial safety

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Nanotechnology has the potential to solve many of modern society's problems, notably in medicine and the environment. Although an increase in interest in regulating risks from nanotechnologies is evident in recent years, research and regulation of risks lag behind the driving forces of intellectual curiosity and commercial potential. The aim of this work is to contribute to a better understanding of the risks of nanotechnology and ultimately to their control. We specifically focus on the reactivity and the potential toxicity of HAP NPs, which are used in many medical innovations.

A variety of HAP NPs were synthesised selected on the basis of their biocompatibility and particle size. Full detailed physicochemical characterisation was carried out in parallel. The synthesis method was then optimised to provide control over crystal growth, agglomeration, particle size and shape mainly by investigating the effect of synthesis parameters such as: synthesis temperature, pH, maturation time, concentration of the capping agent, drying and calcination temperature.

The reactivity/chemical stability of the nanoHAPs was assessed by solubility experiments, which showed that solubility exponentially increases as the grain size decreases. This is in agreement with the modified version of the Kelvin equation, which describes the prediction that solubility is dependent on the particle size and is expected to increase exponentially as particles get smaller. Furthermore, these experiments showed that solubility correlates well with the crystallinity of nanoHAPs.

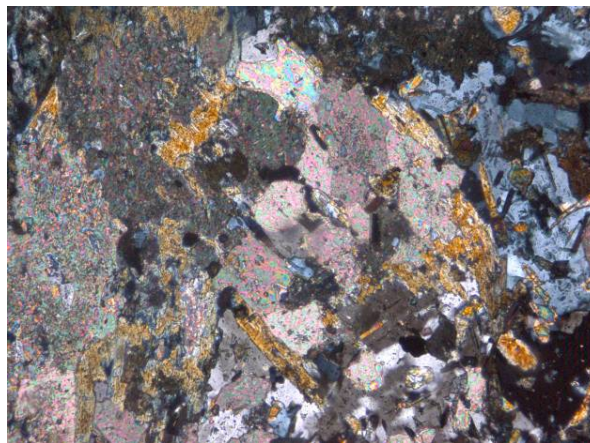
Protein adsorption studies were carried out as a proxy for toxicity; high proportion of protein adsorbed indicates better biocompatibility, and therefore less likelihood for toxicity. These experiments showed that more protein was absorbed by the smallest, least crystallised and non-functionalised HAP NPs, suggesting that these are the most biocompatible nanoHAPs.

## Flux rates for water and carbon during greenschist facies metamorphism

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The time-averaged flux rate for a CO<sub>2</sub>-bearing hydrous fluid during greenschist facies regional metamorphism was estimated to  $10^{-10.2 \pm 0.4} \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ . This was evaluated by combining 1) Peclet numbers obtained by chromatographic analysis of the propagation of reaction fronts in 33 metamorphosed basaltic sills in the SW Scottish Highlands (Fig. 1), 2) empirical diffusion rates for CO<sub>2</sub> in water obtained by Wark & Watson (2003), and 3) calculated time-averaged metamorphic porosities. The latter were calculated using an expression obtained by combining estimated Peclet numbers with the empirical porosity – permeability relationships obtained by Wark and Watson (1998) and Price *et al.* (2006) and Darcy's law. This approach yielded a time-averaged metamorphic porosity of  $10^{-2.6 \pm 0.2}$  for greenschist facies conditions. The corresponding timescale for metamorphic fluid flow was  $10^{3.6 \pm 0.1}$  years. By using mineral assemblages to constrain fluid compositions, I further obtained a time-averaged annual flux rate for carbon of 0.5-7 mol-C. m<sup>-2</sup>. yr<sup>-1</sup>. This matches measured emission rates for metamorphic CO<sub>2</sub> from orogenic hot springs. These fluxes significantly exceed estimated rates of CO<sub>2</sub> drawdown by orogenic silicate weathering and therefore indicate that orogenesis is a source rather than a sink of atmospheric CO<sub>2</sub>.



**Figure 1:** Photomicrograph (in XPL) showing replacement of amphibole by calcite: evidence of carbon transfer, preserved in a metabasaltic sill from the SW Scottish Highlands.