Retention of colloids at rough mineral surfaces: An analog study

GOPALA KRISHNA DARBHA¹, ALEX MICHLER¹, THORSTEN SCHÄFER², ANDREAS LÜTTGE³ AND CORNELIUS FISCHER^{1,3}

¹Georg-August-Univ., Göttinigen, Germany ²INE, KIT, Karlsruhe, Germany ³Rice Univ., Houston, TX, USA

As an analog to mineral surfaces with halfpores in the submicron size we studied the deposition behavior of latex colloids on a well-defined, structured oxidized silicon wafer surface. The topography consists of a regular pit pattern (pit diameter=400 nm, pit spacing=400 nm, pit depth=100 nm). Effects of hydrodynamics and colloidal interactions in transport and deposition dynamics of colloidal particles under laminar flow conditions were investigated in a parallel plate flow chamber. The experiments were conducted at pH ~6.8 under conditions of electrostatic repulsion for negatively charged colloids with plain and carboxylate functionality to study the impact of surface topography on particle retention. Vertical Scanning Interferometry (VSI) was applied for surface topography characterization and the quantification of colloidal retention over scan areas of ~8100 μ m². Despite electrostatically unfavorable conditions, a significant retention of plain and carboxylate colloids was observed at the rough substratum, governed by van der Waals attraction.

The influence of particle diameter variation on retention of monodisperse (from the range of 300-4000 nm) as well as polydisperse (300-1000 nm vs. 1000-4000 nm) suspensions, was studied, also as a function of flow rate over a wide range (Peclet number = 10^{-4} to 1.0). According to filtration theory [1], for larger particles (>2000 nm, e.g. size range of clay mineral aggregates) a linear correlation between the diameter of adsorbed particles and flow velocity was found. Smaller particles (<2000 nm, e.g. size range of clay minerals and iron oxide aggregates) and polydisperse suspensions show, however, an inhomogeneous behavior of deposition rate with increasing fluid flow velocity (Peclet number). At a given flow velocity (where pe<1), for monodisperse colloids, there was no 'shadow effect' [2, 3] observed around particles. In contrast, a polydisperse particle suspension (pe>1) shows significant prevention of further adsorption at a defined area around the particle. This study highlights the importance of colloidal size and polydispersity for the prediction of particle retention at rough mineral and rock surfaces in the environment under unfavorable conditions [4].

[1] Tufenkji et al. (2004) Env Sci Tec 38, 529. [2] Ko et al.
(2000) Env Sci Tec 34, 3681. [3] Loenhout et al. (2009) Coll Surf A. 342, 46. [4] Darbha et al. (2010) Langmuir (accepted).

Evaluating the source and pathway of Pb incorporation in human bone and white-tailed deer through the use of Pb isotopes

THOMAS H. DARRAH¹, ROBERT J. POREDA², JENNIFER J. PRUTSMAN-PFEIFFER³ AND ROBYN E. HANNIGAN¹

 ¹Environmental, Earth and Ocean Sciences, University of Massachusetts-Boston, Boston, MA 02125 USA
 ²Earth and Environmental Sciences, University of Rochester,

Rochester, NY 14627, USA

³Department of Pathology, University of Rochester Medical Center, Rochester, NY 14627

We analyze stable lead (Pb) isotopic composition (²⁰⁷Pb/²⁰⁶Pb, ²⁰⁸Pb/²⁰⁶Pb) and Pb concentration of human cortical and trabecular bone to evalute the source of Pb incorporated into biominerals. Pb isotopic analysis was performed by a combination of TIMS and solution ICP-MS. The isotopic composition of biominerals is expected to reflect the isotopic composition of blood and extracellular fluid at the time of biomineral formation. As a result, Pb isotopic composition may serve as a proxy for the source and changes in the source (e.g. food, water, aerosols, work place exposure) of Pb incorporation throughout time. We present a comparison of human bone tissue Pb isotopic composition between modern Rochester, NY bone and Rochester, NY area archaeological bone dating back to the 1750's. We compare bone composition to Rochester soils (reflecting a mixture of anthropogenic and natural inputs), and lake sediment cores (including both modern and pre-anthropogenic sections), to evaluate changes in the source of Pb incorporated into the body. Additionally, we compare the Pb isotopic composition of a high resolution (0.25cm interval) PA sediment core profile located near a coal fired powerplant to the composition observed in PA deer antlers from the same reservoir from 1974-2007 to evaluate the relative proportion of Pb incorporated from aerosols inhaled and/or deposited and subsequently ingested. Archaeological Pb concentrations and isotopic composition are similar to natural geological inputs and reflect higher total Pb incorporation. While the total Pb concentration has decreased with time, the Pb isotopic composition of modern Rochester bone and white-tailed deer antlers suggests a change towards a significant contribution of Pb from local aerosols inputs.