Migration of PEG-Functionalized Model Gold Nanoparticles in Natural Barriers

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For the design and safety assessment of high level waste (HLW) deep geological repositories, the understanding of radionuclides migration is essential. The migration of radionuclides when colloidal materials or nanoparticles are present has become of high interest due to their transport and mobility enhancement.

In this sense, transport experiments using colloids or nanoparticles are necessary for a more complete understanding of radionuclides mobility and the associated transport mechanisms. In this work, as a model or representative colloidal material, gold nanoparticles (AuNPs) of different Au-core sizes (10, 50 and 100 nm) with their surfaces functionalized with polymeric poly(ethylene) glycol (PEG) chains (**Fig. 1**) where used in column and batch experiments with silica sand, to understand the migration behavior and the transport controlling mechanisms. The obtained breakthrough curves (BTCs) showed asymmetrical curves with dynamic sloped or inclined plateau regions accompanied by retention for the AuNPs, while the HTO tracer showed a typical behavior (**Fig. 2**).

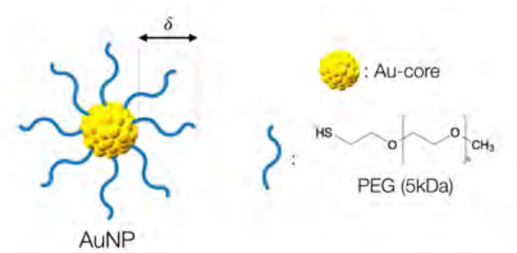

Figure 1. AuNPs schematic representation (left) and legend (right), where δ is PEG thickness.

Dynamic sloping plateaus occur when the negatively charged AuNPs deposit onto positive sites possibly due to accessory minerals followed by interactions and adsorption onto deposited AuNPs by approaching AuNPs, known as ripening.

Moreover, temporal evolution studies of the column effluents by UV-vis spectroscopy indicated aggregation of 50 and 100nm AuNPs. The aggregations in the effluents were possibly caused by the interactions occurring between the deposited AuNPs with the incoming AuNPs within the porespace until detachment starts to occur. Aggregates of 50 and 100nm were observed near cracks and fractured regions on silica sand surfaces from electron microscopy studies at high magnifications, whereas 10 nm AuNPs were found unaggregated in smooth areas. The differences in the migration behavior of different sized AuNPs may be attributed to the differences in the PEG steric stabilizing effects (higher when δ is long) which reduces surface interactions rather than electrostatic stabilizing effects.

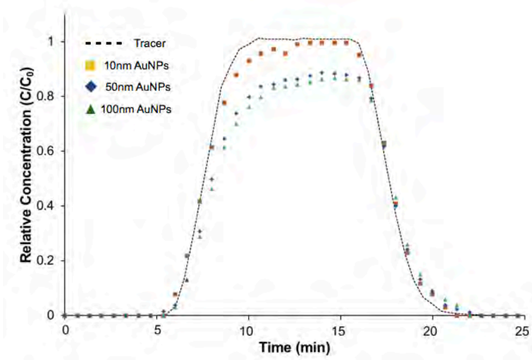

Figure 2. Breakthrough curves (BTCs) for AuNPs pulse injection in quartz-sand at a flow rate of 1.5mL/min. Data is displayed as normalized effluent concentration (C/C_0) as a function of time.