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学　位　論　文　内　容　の　要　旨

博士の専攻分野の名称　　博士（工学）　　氏名　Carlos Eduardo Ordonez Castillo

学　位　論　文　題　名

Application of gold nanoparticles as representative colloids to understand environmental fate and behavior in barrier materials relevant to radioactive waste disposal
(金ナノ粒子を適用した放射性廃棄物処分バリア材中のコロイドの環境動態・挙動の研究)

Studies on environmental fate and transport of colloidal materials through engineered and natural barriers are highly important for the safety assessment of radioactive waste repositories since the migration behaviors of radionuclides may be significantly affected by interactions with colloids. Synthesized gold nanoparticles of various sizes and surface properties can be used as model colloidal materials to study their migration behaviors in barrier systems to understand the effects of the properties of gold nanoparticles on their migration behaviors. In this study, gold nanoparticles (AuNPs) of three different sizes (10 nm, 50 nm, and 100 nm) coated with polyethylene glycol (PEG) were used to study the interactions with and migration behavior in silica sand. In addition, interactions of AuNPs with montmorillonite, a typical engineered barrier material to be used in geological disposal, were investigated. The results suggest that there are cases that neither electrostatic nor van der Waals (vdW) interactions control the migration behavior of AuNPs, and other interactions including steric effects arising from the surface coating need to be considered when assessing fate and transport of nanoparticles.

In Chapter 1, the need for proper radioactive waste management is reviewed as background, including the concepts for near-surface and deep geological repositories of radioactive waste, with an emphasis on the importance of performance or safety assessment of repositories to include colloid mediated transport. Colloids are defined, and their origins are presented as well as their interaction behavior and the available mathematical models that try to explain them. The complexities and limitations for evaluating natural systems and colloids limit the understanding of the impact of colloid mediated transport. Thus, the potential application of AuNPs as representative natural colloids is proposed, their synthesis, properties and their surface functionalization are presented. The unique size and physicochemical properties found in AuNPs allows an easy characterization and detection that can enhance the current understanding of the interactions between colloidal materials and geomedia that can influence colloidal migration.

In Chapter 2, transport experiments using three different sizes of polyethylene glycol (PEG) coated AuNPs were carried out in a porous media composed of silica sand to obtain breakthrough curves. The model AuNPs were characterized by size (in solution and dry), surface charge, surface coating length and optical properties (absorption spectra). The silica sand porous media was also characterized in size, morphology and in chemical composition. Batch experiments were carried out to understand the AuNPs colloidal stability and interactions with silica sand. The absorbance spectra of each AuNPs were used to monitor changes in the concentration and in the properties of the AuNPs including aggregation. The transport of AuNPs was observed to be increased for the smaller nanoparticles, and

the batch experiments showed that the smaller particles exhibit increased stability against aggregation, although DLVO theory predicts more mobility and stability for larger particles. These results suggest interactions other than electrostatic and vdW, including steric effects provided by the PEG coating can control the transport behavior in porous media.

In Chapter 3, interactions of PEG-coated AuNPs of three different sizes with silica sand grains were studied in detail. AuNPs were deposited onto silica sand grains in batch experiments, and samples were randomly extracted, vacuum dried and prepared for scanning electron microscopy (SEM) imaging by coating them with carbon. Elemental mapping was performed to confirm the local chemical composition of the silica sand in order to relate it to the presence of AuNPs. The attachment behavior of the AuNPs observed in the SEM images were consistent with the results from the column transport experiments, where the absorption spectra indicated that aggregation occurred and that it was likely the reason for increased AuNPs column retention. The deposition of AuNPs was observed to be more frequent in the rough surfaces of the silica sand particles, where accessory minerals (surface heterogeneities) are present and that can interact electrostatically with the AuNPs and produce aggregation depending on the nature of the interactions. A transport mechanism for the different AuNPs was proposed based on the results of the transport and batch experimental results.

In Chapter 4, the interactions between different sized PEG-coated AuNPs and Na- montmorillonite were studied in batch experiments varying AuNP concentrations and pH. The attachment and/or deposition behavior of different AuNPs on Na-montmorillonite particles were examined by UV-Vis, TEM, SEM and EDS. The results obtained from the absorption spectra showed that possible interactions between the AuNPs and Na-montmorillonite particles. The results of SEM observations showed that no preferential attachment of AuNPs on to edges or surface of Na-montmorillonite, contradicting with previous reports of and colloidal theory that showed that preferential attachment would occur on the edges due to the presence of surface charge heterogeneities that allow attractive interactions with the AuNPs. This also suggests a possibility of alternative interactions that can control and modify the attachment behavior of the AuNPs such as steric effects.

In Chapter 5, the main conclusions and most relevant findings of this work are presented and summarized.