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Effects of flow interruption on transport and retention of iron oxide colloids in quartz sand



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G R A P H I C A L A B S T R A C T



HIGHLIGHTS

- Iron oxide colloids are highly mobile in quartz sand under continuous flow.
- Colloid retention increases with the duration of flow interruption phases.
- After 112 h of flow interruption, almost no colloids can be released by rinsing.
- Retention is likely related to locally attractive DLVO/XDLVO interactions.
- A mechanistic model is capable of describing the experimental results.

A R T I C L E I N F O

ABSTRACT

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Abbreviations: OMCG colloid, organic matter coated goethite colloid; XDLVO, extended DLVO approach.

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was investigated under continuous and stagnant flow conditions. Classic DLVO and extended DLVO (XDLVO) interaction energies including Lewis acid/base parameters were evaluated using measurements of sessile drop contact angles and zeta potentials of OMCG colloids and quartz. Results elucidated that under continuous flow conditions, OMCG colloids were highly mobile, which was in agreement with calculated unfavorable attachment conditions revealed by predictions of both DLVO approaches. In contrast, during intervals of flow interruption, significant amounts of OMCG colloids were retained in the solid matrix and could not be remobilized via re-establishment of flow. The magnitude of colloid retention increased with the duration of flow interruption; OMCG colloids were almost completely immobilized after 112 h. Further experiments were conducted in order to determine possible colloid retention mechanisms. Results indicated that the major cause for retention during flow stagnation was OMCG colloid capture at locations with attractive DLVO/XDLVO interactions, promoted by fast gravitational settling of colloids onto the solid matrix. We compared breakthrough curves to model predictions, where we demonstrated that an attachment term with a stagnant fluid switch was required in the mass balance in order to reproduce the measurements. We conclude that high mobility of OMCG colloids and prediction of that transport behavior with the applied DLVO approaches were only valid under continuous flow conditions. Under more discontinuous hydraulic conditions relevant in natural soils, such as flow interruption, OMCG colloid transport behavior was modified significantly.

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1. Introduction

In soils and sediments, mobile colloidal iron oxide particles are ubiquitous [1–7] and play an important role in the cycling of iron in the environment [8]. The mobility of iron oxide colloids has received a significant amount of attention in the field of contaminant transport studies, as the colloids can facilitate co-transport of adsorbed metals and radionuclides [1,2,6,9–12]. Moreover, iron oxides and zerovalent iron, which is encapsulated in a shell of iron oxides under environmental conditions, show promising potential in remediation efforts [13,14].

Below their point of zero charge (pzc) of pH 7–9 [15–17], iron oxide colloids can be rapidly immobilized [8–11] due to attractive electrostatic interactions with the mostly negatively charged soil matrix minerals, e.g. quartz [18,19]. However, negatively charged surface coatings can lead to high iron oxide colloid mobility below their pzc [10,11,20–23]. This can be explained with the DLVO approach, which consists of van der Waals and electrostatic interactions and predicts an energy barrier between colloids and matrix [21,23,24]. The XDLVO approach is extended with Lewis acid/base interactions to include effects of surface wettability. It has been applied for clay minerals [25] and model microspheres [26–28], but not yet for iron oxide colloids.

The studies reporting high mobility of negatively charged iron oxide colloids cited above have been conducted under continuous flow conditions. However, the complex flow regimes in natural soils involve phases of flow interruption that should be taken into consideration.

In general, phases of flow interruption lead to replenishment of the colloidal particle storage in soils [29]. The effect of flow interruption on iron oxide colloid mobility has been regarded under reducing conditions. The production of dissolved ferrous iron by reductive dissolution of iron oxides increased with the duration of flow interruption. This causes higher iron oxide colloid formation upon re-establishment of flow and transport of the solution into oxidizing regions [30]. An increase of colloidal iron subsequent to flow stagnation in the effluent of natural soil under reducing conditions was likewise reported by Zhu et al. [31].

There has been no research on the effects of flow interruption on transport and retention of iron oxide colloids under oxidizing conditions yet. We hypothesize that flow interruption can cause colloid retention by several different processes. See Fig. 1 for a graphic representation of the potential retention mechanisms for negatively charged organic matter coated goethite (OMCG) colloids in a negatively charged quartz sand matrix.

It is known that above a certain colloid/collector size ratio, colloids are subjected to straining in pore throats too small for passage [32–34]. During phases of flow interruption, OMCG colloids may form aggregates, which could result in straining (labeled as "1" in Fig. 1).

Moreover, colloids are subjected to slow transport processes such as gravitational settling or diffusion under stagnant flow conditions [35–37], so that they can get into the vicinity of the solid matrix. This may lead to colloid retention by the following mechanisms:

- 1. Previous studies conducted under continuous flow reported that colloids can be retained in immobile flow zones [38–40]. OMCG colloids may enter such zones during flow interruption and remain trapped there upon re-establishment of flow (labeled as "2a" in Fig. 1).
- 2. Under overall unfavorable attachment conditions, colloid retention can occur at locations with attractive DLVO/XDLVO interactions, i.e. secondary minima outwards of energy barriers [23,26,28,41-46] and heterodomains on the solid matrix where the energy barrier is either absent or sufficiently weakened to allow colloid diffusion over them. Heterodomains can originate from surface charge heterogeneity [46–56] as well as from straining related to surface roughness and grain to grain contact zones [35,46,53-59]. Colloid retention is increased at microscale surface roughness locations due to increasing lever arms for attachment and simultaneously decreasing lever arms of hydrodynamics [53–56]. In contrast, retention is decreased by nanoscale surface roughness, which causes shallow primary minima [55,56]. During flow interruption, OMCG colloids may reach locations with attractive DLVO/XDLVO interactions via sedimentation or diffusion (labeled as "2b" in Fig. 1).

The central objectives of this study were (i) to determine OMCG colloid and quartz sand characteristics in context of the DLVO/XDLVO approaches, (ii) to examine the effect of flow interruption on transport and retention of OMCG colloids in column experiments, (iii) in case colloid retention occurs, to identify the responsible retention mechanisms, and (iv) to formulate a mechanistic model describing the mobility of OMCG colloids as affected by flow interruption. Download English Version:

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