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Model studies for flocculation of sand-clay mixtures

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ABSTRACT

Results are presented from a combined experimental and numerical study aimed at comparing the flocculation behaviour of purely-cohesive (clay) and mixed (sand-clay) sediment suspensions under equivalent controlled hydrodynamic conditions. The experiments were conducted in a grid-stirred settling column and focussed on measuring the parametric influences of grid-generated shear rate and local suspended sediment concentrations on the time-evolution of the micro- and macrofloc size distributions generated in the column, as well as representative maximal and root-mean-square floc sizes. The results indicate that for kaolin clay suspensions under low-medium shear rates, initial aggregation rates and the peak or quasi-equilibrium floc sizes attained increase with the clay input concentration; this latter effect due to the larger proportion of macroflocs generated within these runs. By contrast, under high shear rates, representative floc sizes for kaolin clay suspensions remain largely unchanged over the experimental duration, with little influence from clay input or in-situ concentrations, and no macroflocs present in the resulting floc size distributions. The addition of the fine sand fraction to the kaolin clay suspensions is shown to reduce both initial aggregation rates and the representative floc sizes attained in the column for runs under low-medium shear rates, whilst having negligible effect on the flocculation behaviour for the sand-clay mixtures under high shear rates. These results suggest that the sand fraction inhibits flocculation at lower shear rates due to an additional floc break-up mechanism resulting from direct sand-clay interactions (e.g. particle-floc collisions). The importance of these inter-fractional (sand-clay) interactions diminishes, in comparison to shear-induced floc break-up, under higher shear conditions. A one dimensional vertical (1DV) model incorporating a population balance equation (PBE) that includes new representation of these multi-fractional (sand-clay) collisions is applied to simulate the kaolin clay and sand-clay settling column tests. In general, the 1DV PBE model predictions provide good agreement with the measured in-situ concentrations and quasi-equilibrium floc sizes attained, but under-predict floc sizes during the initial aggregation phase due to uncertainty with the upper boundary condition in the 1DV model domain. Furthermore, the reliance of the 1DV PBE model predictions on empirical floc break-up rates associated with shear-induced floc fragmentation and multi-fractional (sand-clay) collisions warrants further attention to better define the microscale dynamics of these processes for their improved representation in the PBE model. It is anticipated that this multi-fractional approach represents an improved basis for modelling flocculation processes within natural sedimentary environments, such as estuaries and tidal inlets, where bed sediments often consist of interacting cohesive (i.e. muds) and non-cohesive (i.e. silts, sands) fractions.

1. Introduction

Natural sedimentary environments such as estuaries are amongst the most dynamic of coastal zones due to strong variability in sediment fluxes generated by natural hydrodynamic forcing from tidal currents and/or waves, as well as anthropogenic interventions such as dredging operations, port and harbor developments, coastal structures and energy installations. Subtidal and intertidal sediments within these environments typically consist of cohesive muds (e.g. clays and organic matter) and non-cohesive fractions (e.g. fine sands and silts), which can vary significantly in proportion both spatially and temporally (Uncles et al., 2006). The nature of interactions that exist between cohesive and non-cohesive sediments has important implications for modelling transport processes such as flocculation, settling, deposition, erosion and consolidation (Cuthbertson et al., 2008, 2010, 2016) and, ultimately, morphodynamic evolution within estuaries. While the physical behaviour of mixed

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(sand-mud) sediments is known to be significantly different from that of the individual fractions (Whitehouse et al., 2000), many previous studies (Williamson, 1991; Chesher and Ockenden, 1997; van Ledden, 2002; van Ledden, 2003; Waeles et al., 2007; Sanford, 2008; Le Hir et al., 2011) have considered estuaries and tidal inlets as predominantly segregational environments, where the mud and sand fractions are treated largely as independent, non-interacting regimes. As such, there is an acknowledged lack of quantitative information on the key parametric influences on sand-mud interactions in suspension, and their effects on flocculation and settling within mixed sedimentary systems (Manning et al., 2013).

For purely cohesive sediments, flocculation (i.e. particle aggregation and break-up) is known to result from inter-particle collisions due to Brownian motion, turbulent mixing and differential settling (Burban et al., 1989; Dyer and Manning, 1999; Winterwerp, 2002). Many flocculation models for cohesive suspensions are based on the hierarchical "order of aggregation" approach (Krone, 1963), whereby primary mud particles agglomerate to form small, dense microflocs (i.e. $D_f \leq \sim 150$ μm), which in turn can flocculate to form larger, more fragile, macroflocs ($D_f \geq \sim 150$ μm) (Eisma, 1986; Manning, 2001). It is generally accepted that for a given primary particle concentration, aggregation rates and, hence, floc sizes will tend to increase with the turbulence intensity due to an increased number of inter-particle collisions. However, the same turbulent motions also generate shear stresses that can limit floc growth due to disruption and break-up of macroflocs (Winterwerp, 2002; Milligan and Hill, 1998). Most previous laboratory and in-situ field investigations have shown a strong inverse relationship exists between measured floc sizes and turbulent shear rates (Cuthbertson et al., 2010; Burban et al., 1989; Milligan and Hill, 1998; Van Leussen, 1988; Krank and Milligan, 1992; Spicer and Pratsinis, 1996; Manning and Dyer, 1999; Verney et al., 2011; Keyvani and Strom, 2014); only at very low shear rates (i.e. $G < 35\ s^{-1}$ (Mietta et al., 2010)), where aggregation dominates over break-up processes, has floc growth been observed with increasing turbulent shear rates.

Previous flocculation research also indicates that the influence of particle concentration is not as straightforward as conventional wisdom or common assumptions on aggregation behaviour may suggest (i.e. higher concentrations → increased inter-particle collisions → higher aggregation rates and increased floc sizes) (Hill, 1998). While some experimental studies (Oles, 1992; Eisma and Li, 1993) support this hypothesis, other investigations (Cuthbertson et al., 2010; Milligan and Hill, 1998; ten Brinke, 1994) found limited or no systematic correlation between floc sizes generated and mass concentration. Indeed, the study by Tsai et al. (1987). indicated the opposite trend, with reduced floc sizes attained at higher particle mass concentrations, leading Lick and Lick (1988) to suggest that, under specific parametric conditions, inter-floc collisions may be more important in the break-up process than fluid shearing. The temporal variability in these aggregation and break-up processes also results in continually-evolving floc properties (e.g. floc size and density), non-equilibrium suspended sediment loads and settling fluxes (Cuthbertson et al., 2010; Manning and Dyer, 1999; Gratiot and Manning, 2004). Cuthbertson et al. (2010). conducted grid-stirred settling column experiments to investigate the temporal evolution of kaolin clay flocs forming under a range of turbulent shear rates and "single-shot" particle mass concentrations. They found that the highest initial aggregation rates and largest (macro-) flocs were generated under low turbulent shear conditions, with lower aggregation rates and smaller (micro-) flocs generated under higher turbulent shear conditions. However, for the time-varying conditions generated by the "single-shot" clay input, the observed flocculation behaviour was largely controlled by resulting concentration gradients in the column that influenced both the floc settling rates and flocculation time scales (i.e. time to attain the so-called *equilibrium* floc size, where aggregation and break-up processes balance (Winterwerp, 1998)).

In comparison to these extensive studies on purely cohesive (mud) flocculation, there remain significant knowledge gaps in the physical understanding of flocculation and settling behaviour for sand-mud mixtures. Recent experimental studies (Manning et al., 2010, 2011) found that the contributions to the mass settling flux from microflocs (i.e. $D_f \leq 160$ μm) and macroflocs (i.e. $D_f \geq 160$ μm) tend to increase and reduce, respectively, with increasing sand content, leading to the hypothesis that the sand grains tend to interact more with the smaller, denser microfloc structures than with the larger, more fragile macroflocs. By contrast, Cuthbertson et al. (2010). demonstrated that the aggregation rates and floc sizes generated in pure kaolin clay suspensions under non-equilibrium conditions were inhibited significantly by the addition of sand. It is suggested that this demonstrates the role of (i) direct collisions between sand particles and clay macroflocs, and/or (ii) indirect particle-fluid interactions and self-induced turbulence within mixed (sand-clay) suspensions, in increasing the break-up rate of macroflocs, and thus inhibiting their formation in the mixed (sand-clay) suspensions. However, as there have been relatively few systematic experimental studies aimed at determining the parametric influences on the flocculation of sand-mud mixtures, the influence of inter-fractional interactions on aggregation and break-up processes remains largely ignored in flocculation modelling studies.

The main aim of the current study is therefore to investigate experimentally the parametric controls on the flocculation behaviour of both purely cohesive (kaolin clay) and mixed (sand-clay) suspensions for prescribed sediment feed conditions and controlled hydrodynamic forcing generated in a grid-stirred settling column. Specifically, the clay and sand fractions are added (where applicable) continuously over a specified feed duration, with the resulting temporally-increasing mass concentrations in the settling column designed to replicate, to some extent, the increases in suspended concentrations that typically occur during the build up to an estuarine turbidity maximum (Manning et al., 2006). The study extends on from the previous experimental work by Cuthbertson et al. (2010). by measuring the temporal evolution of representative floc sizes (i.e. maximal $D_{f,95}$; $D_{f,90}$ and root-mean-square $D_{f,rms}$) both during the initial aggregation phase (i.e. $dD_f/dt > 0$, under *non-equilibrium* conditions) and the transition to *quasi-equilibrium* flocculation conditions (i.e. $dD_f/dt \approx 0$) within the settling column. Here, the kaolin-only runs are therefore used to provide base-line data against which the parametric influences of the sand fraction in the sand-clay mixture runs could be assessed quantitatively.

The second aim of the study is to simulate these kaolin-only and sand-clay settling column runs through the application of a one-dimensional vertical (1DV) advection-diffusion model coupled to a multi-fractional population balance equation (PBE) approach. A key new aspect of this numerical study is the inclusion in the PBE of additional floc break-up terms to account for the effects of sand particle – clay floc collisions, and determine their influence on floc growth rates and maximum floc sizes attained in the sand-clay mixture runs. To the best of the authors' knowledge, this is the first time that inter-fractional (sand-clay) break-up terms have been included in a 1DV PBE model approach. As such, the experimental datasets from the kaolin-only and sand-clay settling column runs are used to verify the aggregation and break-up terms specified in this new, multi-fractional PBE model, thus enabling the quantitative contributions associated with floc growth (i.e. floc-floc collisions) and break-up (i.e. floc breakage; sand particle – clay floc collisions) to be isolated within the model simulations.

2. Experimental set-up and methodology

2.1. Settling column arrangement

Fig. 1(a) and (b) show a schematic representation of the settling column used in the experimental studies and a labelled image of the system in operation, respectively. The column arrangement consists of a 50 L capacity upper buffer mixing tank; a 2.1 m-long main column section, constructed from circular acrylic pipe with an internal diameter of 0.24 m (5 mm wall thickness); and an *in-situ* floc measurement section at the bottom of the column. For the experimental runs, the column and