Journal of Sedimentary Research

Journal of Sedimentary Research, 2013, v. 83, 1025–1031
Current Ripples
DOI: 10.2110/jsr.2013.77

EXPERIMENTAL DEPOSITION OF CARBONATE MUD FROM MOVING SUSPENSIONS: IMPORTANCE OF FLOCCULATION AND IMPLICATIONS FOR MODERN AND ANCIENT CARBONATE MUD DEPOSITION

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ABSTRACT: **Flume experiments with fine-grained carbonate particles (< 62.5 μm) show that they form floccules that travel in bedload, form current ripples, and deposit laminated sediments. In order to compare our flume experiments to work on sand transport, we ran flume experiments with medium sand and observed that the velocities at which sand grains started to move and form ripples were in the same range as those where floccules move and form ripples. These ripples are in essence identical to those formed by clay-mineral floccules or sand grains under similar conditions. In light of previous experiments with clay minerals, these results indicate that, of the key controls on mud deposition, flocculation and suspended-sediment concentration are more important than particle mineralogy or water chemistry, and are about as important as bottom shear stress for deposition.**

Suspensions of carbonate mud show the same pattern of flocculation, ripple formation, and bed accretion as observed previously in experiments with clay-mineral suspensions. The resulting carbonate mud deposits show internal low-angle (2–5 degrees) laminae, and in plan view a pattern of ripple foresets that is identical to rib-and-furrow structure in sandstones.

Just as previously assumed for terrigenous muds, there has been a long-standing notion that accumulation of abundant carbonate mud reflects quiescent conditions of offshore and deeper-water environments. These experiments demonstrate unequivocally that carbonate muds can also accumulate in energetic settings. In the sedimentary record of carbonate rocks, interbedded grainstones and lime mudstones may thus not necessarily reflect shifts in depositional energy (or water depth), but alternatively may imply a shift in supplied sediment type. The observations we report suggest that published interpretations of ancient lime muds and derived paleoceanographic conditions may need to be reevaluated.

INTRODUCTION

Fine-grained (< 62.5 μm) terrigenous clastic sedimentary rocks, commonly known as shales or mudstones, are the most abundant sedimentary rock type and represent a larger proportion of Earth history than all other sedimentary rock types combined (Schieber 1998). They play a key role in the global carbon cycle and are at the center of the current boom in shale-gas exploration that is transforming global energy markets (Walsh 2011). In spite of their common occurrence and economic importance, however, they are poorly understood when compared with other types of sedimentary rocks.

Notwithstanding the notion that deposition of siliciclastic mud requires quiescent environments (e.g., Potter 2003), recent flume experiments demonstrate mud transport and deposition at current velocities that suffice to transport and deposit sand (Schieber et al. 2007; Schieber and Southard 2009). In these experiments floccules form floccule ripples that accrete into mud beds at flow velocities between 0.1 to 0.3 m/s.

Fine-grained carbonates are common in most carbonate depositional systems (Flügel 2004), and are typically interpreted as products of low depositional energy (e.g., Boggs 2005). Nonetheless, there have been suggestions that, just like terrigenous muds, carbonate muds can flocculate and accumulate in higher-energy environments (Shinn et al. 1989; Shinn et al. 1993).

Our experiments were aimed at examining a set of questions that in our opinion follow logically from above observations. The first and most fundamental question is whether carbonate muds indeed can form floccules. The second question is whether such floccules are strong enough to pass into bedload through the basal shear layer of a moving suspension, and whether moving bedload floccules give rise to ripple formation, analogous to what we observed in clay experiments (Schieber et al. 2007). In addition, an issue that has been examined from various perspectives in carbonate sedimentology for many years is the potential role of organic coatings in the aggregation of carbonate grains (e.g., Boyer 1972; Riding and Awramik 2000; Flügel 2004). Thus, examining the potential role of organic coatings for producing and stabilizing carbonate mud floccules was the third question we wanted to examine.

In this study, we used carbonate mud collected from natural settings to examine the depositional behavior of suspensions of carbonate mud in flume experiments. These experiments followed the same protocol used for our experiments with clay suspensions (Schieber et al. 2007; Schieber 2011a). Moving carbonate mud suspensions formed floccules, ripples, and rippled beds once the flow velocity was below the critical velocity for sedimentation (Schieber 2011a). Although Shinn et al. (1989, 1993) provided observations that suggested carbonate mud flocculation, to our knowledge this is the first study in which flocculation of carbonate mud was observed directly, in which bedload transport of these floccules is

Published Online: November 2013
 1527-1404/13/1025-1031/$03.00

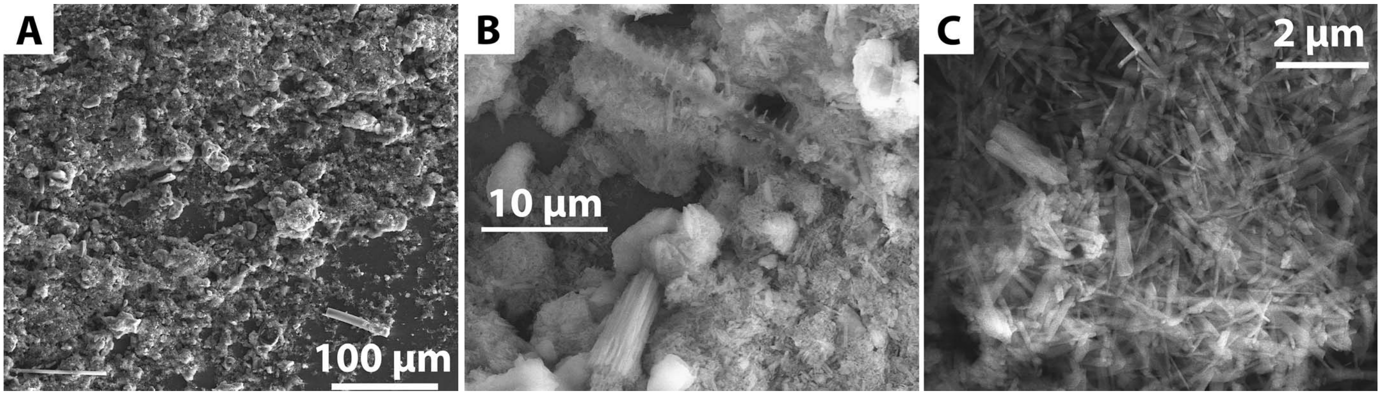

FIG. 1.—SEM images of solid particles in Florida Bay carbonate mud that was sieved to remove particles > 62.5 µm and bleached to remove organic coatings (subsample 1). **A)** Coarse fraction. Abundant particles (clumps) in the tens of micrometers size range. These particles consist largely of calcite. **B)** Fine fraction. A mixture of particles smaller than 10 µm and a groundmass of small aragonite needles. **C)** Close-up of needle-rich groundmass.

demonstrated, and in which the final product (accreted bed of laminated carbonate mud) can be related to bedload transport and migration of floccule ripples. Observations from modern carbonate environments and from the rock record suggest that deposition of carbonate muds by currents could have been common throughout geologic history.

EXPERIMENTAL APPROACH

The experiments were conducted with the same equipment and using the same procedures as in prior experiments with clay suspensions (Schieber et al. 2007; Schieber and Southard 2009). The flume is a racetrack design and uses a flow lid to maintain uniform shear stress along the channel. Flume experiments were monitored via (1) time-lapse photography, (2) filming ripple migration in near-wall situations, and (3) digital high-definition videos of the flume bottom through a macro-lens (10 µm/pixel resolution).

Carbonate mud collected from Florida Bay was passed through a 62.5 micrometer sieve (X-ray diffraction data indicate 60% aragonite and 40% high-Mg calcite) and then stored in a cold room. The Florida Bay mud (FBM) contains abundant needle-like aragonite crystals that range in length from 0.5 to 5 µm (Fig. 1) as well as larger bioclasts and peloids (Flügel 2004). This material, if left at room temperature, develops a strong fetid odor within days due to decay of organic coatings on grain surfaces (e.g., Mayer 1994). Half of the sieved mud was mixed with a 10% sodium hypochlorite solution (bleach) and allowed to react for 1 month (subsample 1), and the other half (subsample 2) was left in its natural state (raw) and stored in a cold room. Scanning electron microscope (SEM) examination showed that organic coatings had effectively been removed from grain surfaces of subsample 1. The reason why we prepared ‘‘bleached’’ and ‘‘raw’’ sample material was to examine the question whether the organic coatings on natural (raw) material significantly affect flocculation, transport, and deposition of carbonate muds.

Grain-size analysis of subsample 1 with a Micromeritics SediGraph showed that more than 65% of the carbonate particles were smaller than 10µm and 90% were smaller than 30 µm. Subsample 1 was multimodal, with a ‘‘coarse’’ mode centered at approximately 35 µm (Fig. 2). The ‘‘coarse’’ mode consists of above-mentioned bioclasts and peloids.

A portion of subsample 1 (bleached FBM) was vigorously stirred in a 40 liter bucket, allowed to settle for 5 minutes, and then the partially settled suspension was decanted. The decanted portion (subsample 3) was ‘‘fines dominated’’ because most of the coarser fraction (20–60 µm) had been removed through settling. Subsample 3 was used in initial flume experiments to demonstrate floccule formation by carbonate fines. Individual floccules were captured on grooved glass slides (Schieber et al. 2007), air dried, and examined by SEM (Fig. 1A, B, C).

For subsamples 1 and 2, identical flume experiments were conducted at room temperature (23 °C) with sediment concentrations that increased stepwise from 0.75 to 3 grams per liter. These experiments followed the same procedures as described in Schieber et al. (2007). Salinity was 35‰, and flow velocities ranged from 15 to 50 cm/s at 5 cm effective flow depth. Reynolds numbers (calculated for flow through a rectangular duct) ranged from 21,450 at 0.15 m/s to 71,500 at 0.50 m/s, and the flow was therefore considered fully turbulent (flow are turbulent when the Reynolds number exceeds 4000). Because the racetrack flumes we used for these experiments operate with a flow lid to keep shear stress uniform along the flume channel (Schieber et al. 2007), we measured the pressure drop along the flume channel to derive values of average shear stress for these experiments. Floccule images were processed with the ImageJ public-domain software package.

OBSERVATIONS

In experiments with ‘‘fines-dominated’’ carbonate mud (subsample 3), formation of abundant large floccules (Fig. 3E) and floccule ripples (Fig. 3D) occurred at flow velocities of 25 cm/s or lower. Unlike sand

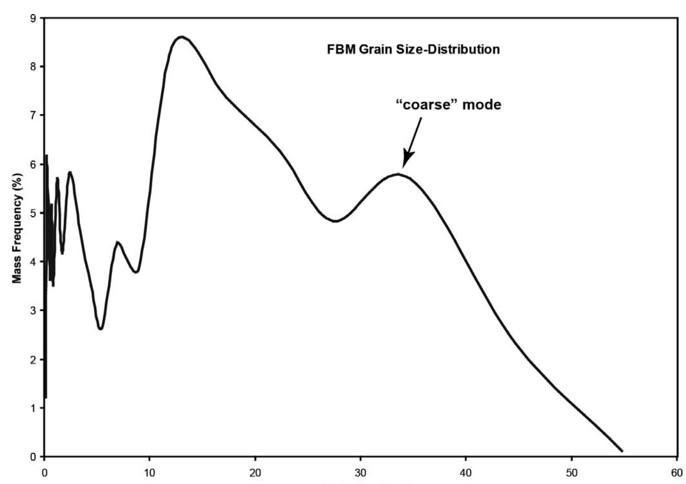

FIG. 2.—Grain-size distribution of sieved (< 62.5 µm) Florida Bay carbonate mud. Note the ‘‘coarse’’ mode between 35 and 40 µm. This material largely forms the ripples visible at velocities between 35 and 50 cm/s.

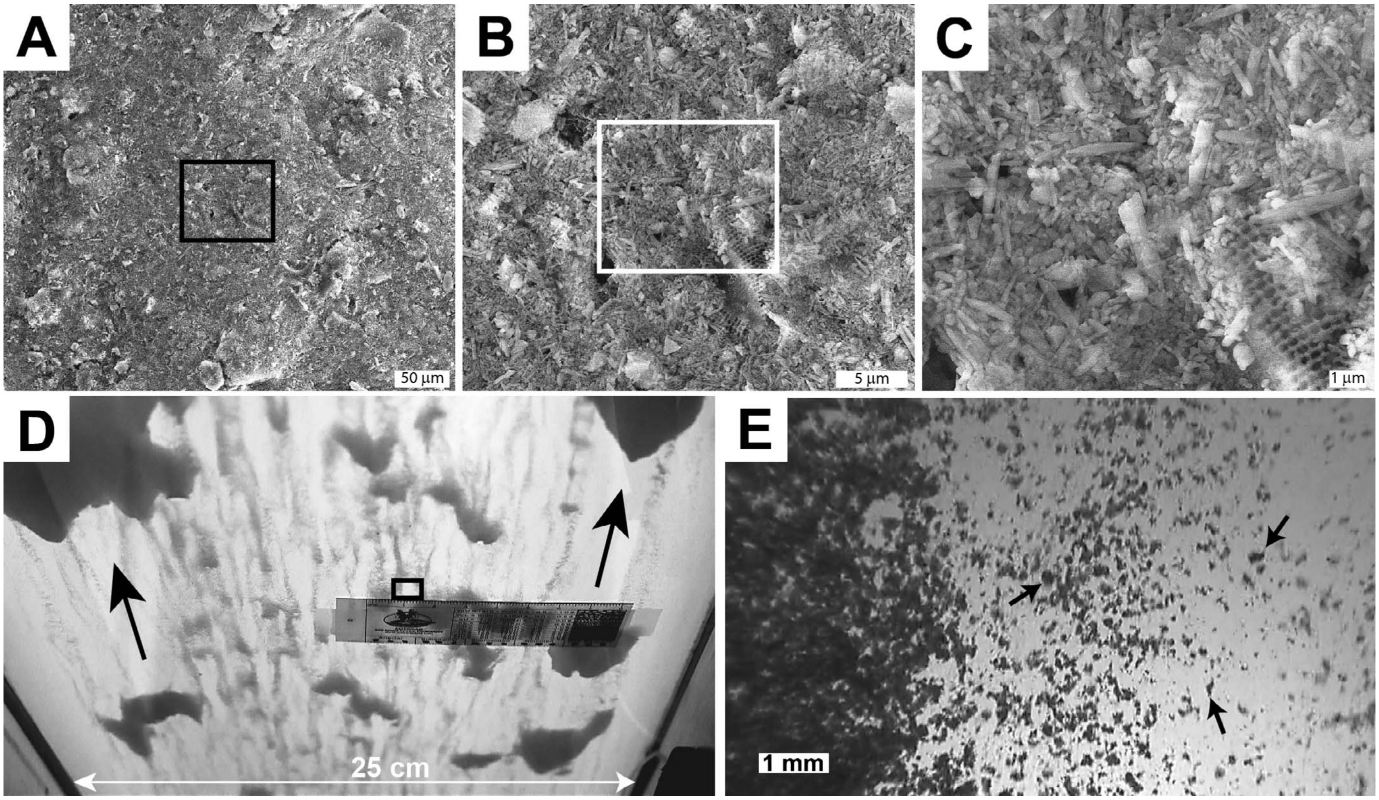

FIG. 3.—A–C) SEM images of floccules that formed from subsample 3 (fines dominated FBM). **A)** center of a captured floccule. Shows abundant fine material and a small proportion of clumps and particles in the tens of micrometers size range. **B)** Close-up of the area marked by black square in Part A. Note abundance of micrometer size needles that enclose larger clumps and fossil debris. **C)** Close-up of fine needle-dominated material that constitutes the bulk of the floccule. **D)** View of flume channel from below (with lighting from above). Flow velocity is 25 cm/s, and ripples of flocculated carbonate mud (as in Part A) have formed. The width of the channel is 25 cm (see white double arrow), and flow direction is indicated by black arrows. The black rectangle in the central region of the image marks the area shown in close-up in Part E. **E)** A high-definition video frame taken at the location of the black rectangle in Part D. At left, floccules have piled up; they are part of a migrating ripple. In the center, individual floccules are clearly visible (examples pointed out with black arrows). Floccules that seem out of focus are either moving too fast for the frame rate of the video camera or are outside the camera's focal plane. Floccules are several hundred micrometers in size.

ripples, the floccule ripples show cohesion between particles due to the intrinsically cohesive nature of floccules, as evidenced by erosion of millimeter-size chunks of ripples during migration. SEM examination of captured floccules (Fig. 3A, B, C) showed them to consist largely of micron-sized aragonite needles. This result demonstrated that micron-size carbonate particles can indeed flocculate and that these floccules form bedload ripples.

In flume experiments with bleached (subsample 1) and raw (subsample 2) Florida Bay carbonate mud there was no discernible difference in the threshold velocities of ripple and floccule formation (Schieber et al. 2007). The latter experiments showed ripple formation across the entire velocity range (15–50 cm/s). At high velocities (35–50 cm/s) ripples had a length-to-width (l/w) ratio between 1 and 2 (Fig. 4A, B). Below 35 cm/s l/w ratios of ripples increased. Ripples formed at 30 cm/s had l/w ratios between 2 and 3 (Fig. 4C), and at 25 cm/s and lower the l/w ratio ranged from 5 to 30 and ripples accreted to form a layer of carbonate mud (Fig. 4D, E).

At flow velocities between 35 and 50 cm/s, flume-bottom video frames showed bedload particle size to be in the 35 to 40 micron range (Fig. 5). At 30 cm/s the mean bedload particle size showed a slight increase (Fig. 5), and upstream portions of ripples showed some adhesion to the flume bottom (Fig. 4D; increase of l/w ratio). Finally, at 25 cm/s, there was a clearly discernible increase of mean bedload particle size (Fig. 5) and analysis of video frames showed that more than 50% of the particles were in the 50 to 200 micron size range (Fig. 5). Mean bedload particle sizes further increased at still smaller flow velocities (Fig. 5).

Observing these ripples from below with back-lighting shows that the lee slope of ripples results from the buildup of successive lobes of flocculated material that move down the lee slopes of the ripples. In experiments with flocculated clays, occasional wall intercepts of floccule ripples (Schieber and Southard 2009) show them to have cross sections comparable to sand ripples. Wall intercepts of ripples formed in these experiments (Fig. 4F) showed a ripple index (length/height) between 15 and 20, within the range that has been reported from sand ripples (e.g., Boersma 1970; Reineck and Singh 1980). Gravimetric analysis of the water content of the final deposits of these experiments immediately after draining the overlying water indicates water contents of 80 to 85 volume percent.

At the conclusion of our carbonate-mud experiments, the final deposits showed ripples and ripple textures at the bed surface (Fig. 6A, D). Mud beds were air dried to a consistency of soft butter and then scraped with a spatula for examination of internal structure. The scrapes revealed downcurrent-dipping low-angle laminae (Fig. 6C) as well as parallel-appearing internal laminae (Fig. 6D). When the sediment was still moist, these laminae varied in color and smoothness (Fig. 6C, D). The darker smooth laminae are dominated by flocculated finer material, whereas the lighter and rougher-appearing (plucked) laminae consist of coarser silty material. In plan view these laminae show rib-and-furrow structure (Fig. 6B).

Ripples composed of fine-grained carbonate material have also been observed by divers on modern carbonate banks (Fig. 7A). One of us

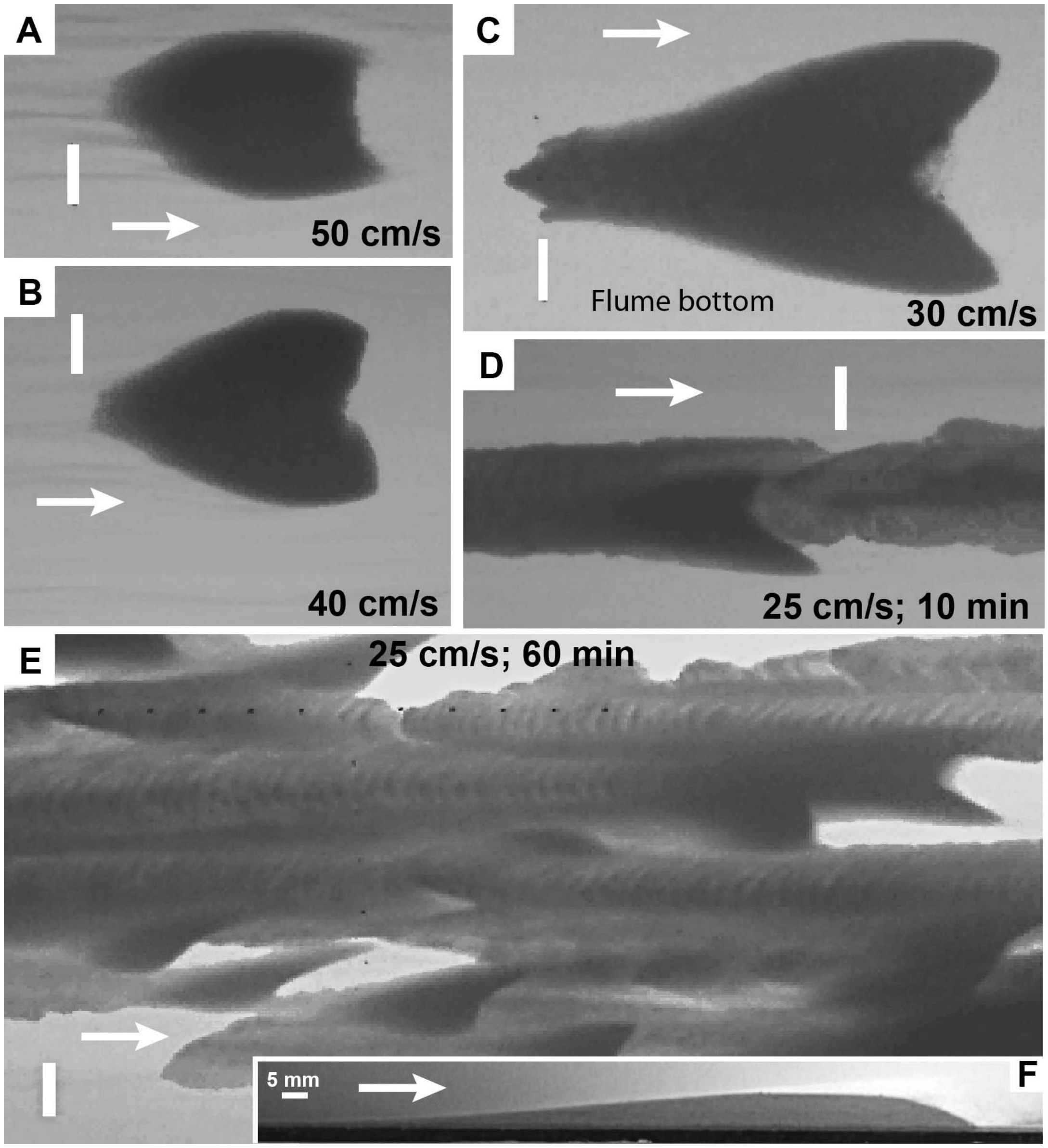

FIG. 4.—Ripple appearance in relation to flow velocity. **A**, **B)** High-flow-velocity ripples with small l/w ratio. **C)** Ripple elongation due to "sticky" cohesive "tails." **D)** Onset of accretion and build-up of ripple "tails" below critical velocity of sedimentation. **E)** A larger and larger proportion of the flume bottom becomes covered with sediment. The sediment is deposited by ripples. Over the coming hours, continued ripple migration will lead to accretion of a bed of carbonate mud. **F)** Cross section of a ripple that intersected the flume wall. Arrow indicates flow direction. Scale bars in Parts A through E are 1 cm in length. The images (A–E) were taken from underneath the flume with back lighting. Light gray areas are the flume bottom without sediment cover.

(RNG) had the opportunity to observe this type of ripples directly, and found them to be asymmetrical like typical current ripples and of low density. They do not offer noticeable resistance to touch and are easily destroyed and dispersed. In these properties they appear very similar to ripples composed of flocculated aragonite in experiments presented here.

DISCUSSION

In earlier experiments on mud deposition from moving clay suspensions (Schieber et al. 2007), flocculation of clays led to ripple formation and bed accretion at flow velocities of approximately 25 cm/s or lower. The observation that we see a comparable behavior in experiments with the "fines-dominated" carbonate mud of subsample 3 suggests that fine-grained carbonates undergo flocculation just as do clays, and that the resultant floccules are of comparable strength. This interpretation is plausible because crystal lattices are repeating patterns of atomic organization, and therefore the edges of all minerals carry unsatisfied charges. At small interparticle distances and small grain sizes these forces from unsatisfied charges (van der Waals forces) are strong enough to induce flocculation (van Olphen 1963) in clay and carbonate minerals alike. The observation that sediment that was bleached (subsample 1) and sediment that still had its natural organic coatings (subsample 2) showed no marked difference in flocculation and ripple formation suggests that the cohesion that is provided via van der Waals forces is of a magnitude similar to that of the "stickiness" caused by organic coatings.

That ripples form at 50 cm/s in subsamples 1 and 2 most likely is due to the "coarse"-mode (~ 35 μm) silt component that is present together with

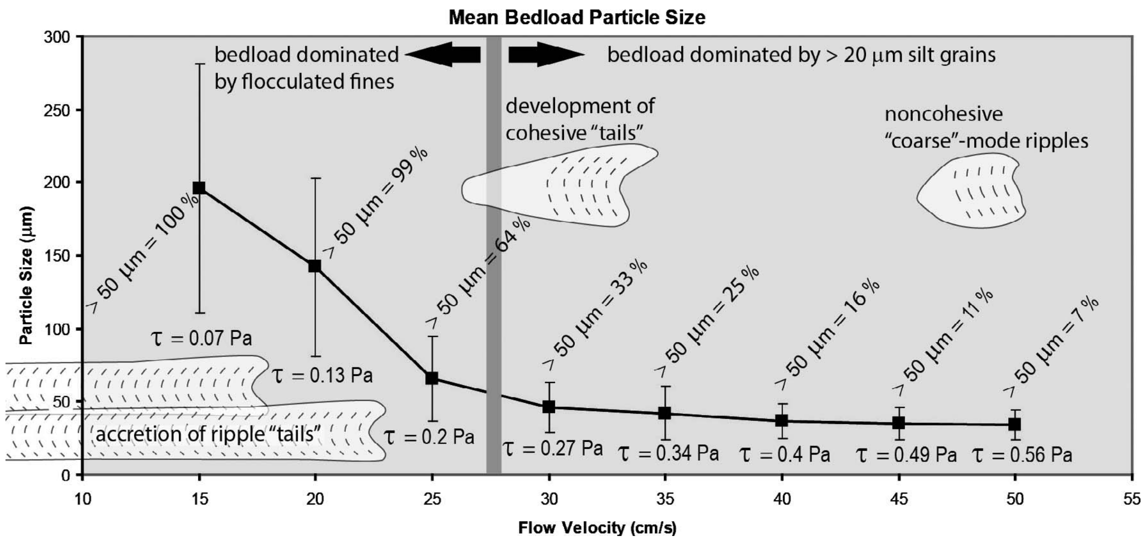

FIG. 5.—Plot of relationships among observed bedload particle size, flow velocity, shear stress τ, percentage of particles above 50 μm in size, and ripple appearance (Fig. 4). Vertical bars denote plus-minus one standard deviation (statistics from 500 floccule size measurements). The diagram shows that at velocities of 30 cm/s or larger, primarily the "coarse"-mode particles are travelling as bedload. Development of cohesive "tails" and ripple elongation becomes increasingly visible at velocities of 40 cm/s and lower. Once the velocity is reduced to 25 cm/s or less, an increasing proportion of flocculated fines appear in the bedload. This happens because at these reduced levels of shear stress the floccules are strong enough to pass through the basal shear layer without being destroyed. Sand-size floccules of fines and nonflocculated "coarse"-mode material travels as bedload at the same time and accretion of ripple tails leads to bed deposition. This data set is from an experiment with 0.75 grams of Florida Bay carbonate mud per liter. Particle size was determined from film frames of material transported across the flume bottom. Shear stresses calculated for 23°C.

the finer, clay-size particles (Fig. 2). Indeed, for flow velocities of 35–50 cm/s the bedload consists largely of "coarse"-mode material (Fig. 5). Prior experimental work with silt-size materials suggests that "coarse"-mode grains should be expected to behave noncohesively and form ripples at these flow velocities (Jopling and Forbes 1979).

Given the grain-size distribution of dispersed FBM (Fig. 2), grains larger than 50 μm are most likely floccules, and the proportion of flocculated material strongly increases below approximately 25 cm/s (Fig. 5). The correspondence among particle size (Fig. 5), formation of floccules (Fig. 3E), l/w ratio of ripples, and adhesion of sediment to the flume bottom suggests that between 25 and 30 cm/s (at shear stresses of around 0.24 Pa) micrometer-size carbonate particles begin to form floccules that are strong enough to pass through the basal shear layer and travel as bedload.

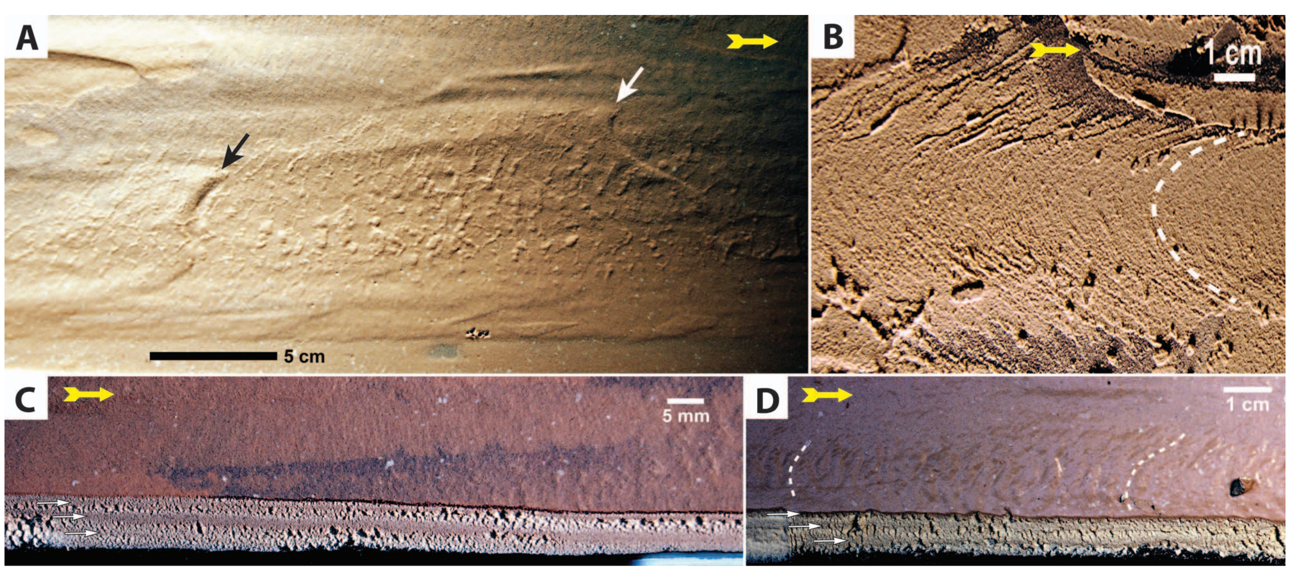

FIG. 6.—Features of flume-deposited carbonate muds (yellow arrows indicate flow direction). **A)** Bed surface after draining the flume. Although somewhat compacted (~ 75% porosity), ripple crests are still visible (black and white arrow). **B)** When the top of the bed is peeled off, the remaining sediment shows rib-and-furrow structure (dashed white line). Parts C and D show internal laminae exposed by scraping the soft sediment. **C)** Low-angle-inclined internal laminae (marked by white arrows). Darker smooth laminae are dominated by flocculated finer material (25 μm and smaller; see Fig. 2), the lighter plucked laminae are dominated by coarse-mode material (25 μm and lager; see Fig. 2). The downlap of these laminae towards the right reflects lateral sediment accretion on the flume bottom. These inclined laminae are not ripple cross-laminae, however, because internally they show rib-and-furrow structure that reflects overlapping compacted ripples. These observations are directly analogous to what has been observed in deposits produced by accretion of clay-floccule ripples (Schieber et al. 2007). **D)** Parallel-appearing internal laminae (marked by white arrows). The curved ridges visible at the surface (dashed lines) reflect the origin of these laminae as the remains of migrating ripples.

Fig. 7.—Modern equivalents of our flume-deposited aragonite muds. The image is of rippled mud from a mud bank in northern Belize (image by RNG). Flow is from the lower left to the upper right; hand for scale.

Because the ''coarse''-mode ripples still move at the velocities that allow the carbonate floccules to travel in bedload as well, ripples composed of coarse silt move over the same surface and at the same time as do floccule ripples. Because the coarse silt grains are much denser than the larger floccules, the coexistence of these particles suggests that they are each other's hydraulic equivalent. Their segregation during transport may in part be due to surface charges, the inherent fragility of floccules, and differences in density and size. Because the coarse silt grains have surface/volume ratios that are one to two orders of magnitude smaller than those of the micrometer-size particles, van der Waals forces between coarse silt grains and the rest of the floccule would be correspondingly weaker. Furthermore, coarse silt grains, even if they were to be incorporated into floccules initially, have as much mass as the rest of the floccule. Thus, every time such a floccule impacts the bed or other particles, the inertia of the coarse silt grain would tend to disrupt and break up the floccule. In addition, bedload floccules are moved much more easily than silt grains because of their lower density (easier to lift) and larger size (larger contact surface). In combination, these factors should ensure effective segregation of coarse silt from floccules during transport. Although the murkiness of the flow does not permit direct observation of these processes, the alternating finer and coarser laminae (Fig. 6C, D) suggest that coarse silt was on the move at the same time as floccule ripples while the bed accreted. Once below the critical velocity of sedimentation (Schieber et al. 2007), the flocculated and cohesive ''fines-dominated'' material that travels in bedload leaves behind thin ''tails'' of sediment (Fig. 4). Stacking and overlapping of these ''tails'' in the course of further ripple migration (Fig. 4E) leads to bed accretion. Because the ''tails'' are the basal portions of ripples that migrate over the surface, they preserve the toe portions of ripple foresets and thus the curved ridges of the rib-and-furrow structures seen in Figure 6B. As silt ripples and floccule ripples successively pass over a given point on the bed, a deposit that consists of stacked up silty and floccule-rich finer laminae results (Fig. 6C, D).

Should the observation of carbonate mud ripples in experiments change the way we look at muddy carbonates? Although accumulation of carbonate mud is commonly thought to reflect low-energy conditions (e.g., Boggs 2005) there have been occasional observations (such as seen in Fig. 7A) that carbonate muds may also accumulate in the presence of wave and current action.

For example, Shinn et al. (1993) report hurricane-deposited laminated lime muds from tidal channels in the Bahamas, and Enos and Perkins (1979) suggest lamination due to wave reworking of carbonate mud for carbonate mud banks in Florida Bay. In addition, Shinn et al. (1989) report on flocculation of carbonate mud in whitings, and note that these sediments formed tiny ripples in water containers due to the rocking of the boat. Similarly, conventional wisdom about low energy requirements for shale deposition discouraged geologists from entertaining high-energy alternatives for many years (Schieber et al. 2007). It is therefore quite possible that more carbonate muds will be interpreted as high-energy deposits once the science behind the concept becomes established.

Lime mudstones are abundant in the rock record (Flügel 2004; Wilson 1975), and quite a few of them may turn out to be of high-energy origin once examined from the perspective that our experiments provide. As far as criteria for recognition in the rock record are concerned, a case for high energy may be difficult to make if we take into account that these sediments will undergo severe compaction (initial water contents ~ 85%) and that parallel-appearing laminae may be the only feature we have to work with. On the other hand, judging from analogous studies in terrigenous clastic mudstones (Schieber et al. 2007; Schieber and Yawar 2009; Schieber 2011a), subtle features like low-angle downlapping of laminae and relict rib-and-furrow structure may still be observable and help make the distinction between carbonate muds that simply settled from suspension and those that were deposited from wave and current action.

CONCLUSION

When comparing flume experiments with carbonate mud with identical experiments that used clay suspensions (Schieber et al. 2007; Schieber and Southard 2009, Schieber and Yawar 2009), very similar behaviors emerge in spite of strongly contrasting mineralogy. Just as in the clay experiments, carbonate mud forms floccules below a critical velocity (Fig. 5), and these floccules form ripples. As these ripples migrate and more floccules form, a mud bed accretes. The resulting carbonate deposits should show parallel and in places low-angle inclined laminae. Upon close inspection, a good portion of laminated lime muds in the rock record may contain features that suggest deposition in the presence of wave and current action.

Until bed shear stresses fall below ~ 0.25 Pa, only the coarser, silt-size, grains travel in bedload as individual particles (noncohesive), whereas the finer, micrometer-size, constituents remain largely in suspension. Once shear stress falls below 0.25 Pa, sand-size floccules that consist of fine, micrometer-size carbonate material (cohesive behavior) can survive transfer through the basal shear layer and add to bedload. At this point the bedload sediment consists of ripples composed of "coarse"-mode particles (coarse silt, Fig. 2) as well as of ripples that are composed of sand-size floccules (Fig. 5), and the accreting bed consists of interspersed silt-rich and floccule-rich laminae (Fig. 6C). Comparable behavior has been observed in experiments that used mixtures of quartz silt and clay (Schieber 2011b), and suggests that in fine-grained sediments, siliciclastic or calcareous, interlamination of silt with clay-size materials is indicative of simultaneous deposition from the same currents, rather than implying strong fluctuations in sediment supply or intermittent current flow and reworking. It also appears that van der Waals forces and organic grain coatings provide comparable degrees of cohesion between small particles. Because the bulk of currently published interpretations of fine-grained laminated sediments propose combinations of low-energy conditions and episodic sediment supply, revisiting these interpretations is likely to lead to a better understanding of the rock record.

ACKNOWLEDGMENTS

The presented research was supported by National Science Foundation grants EAR 0617128 and OCE 0930829. Additional support was provided by the Indiana University Shale Research Consortium, supported by ExxonMobil, Chevron, Anadarko Petroleum Corp., Marathon Oil Comp., and Shell Oil. The manuscript benefited from constructive reviews by M. Tucker, P. Enos, C. Paola, K. Bohacs, C. Soja, and J. Macquaker.

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Received 10 March 2013; accepted 18 August 2013.