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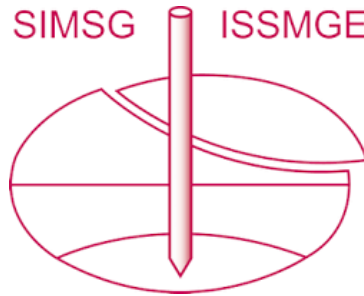
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Understanding the natural consolidation of slurries using colloid science

Comprendre la consolidation naturelle des vases par la science des colloïdes

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ABSTRACT: Many projects (Marker Wadden, Kleirijperij) have recently been initiated in the Netherlands that involve the reuse of dredged material from silted channels. This dredged material is deposited in large ponds, and left to consolidate. When its dewatering and strength development is found to be adequate, the consolidate slurry is used as building or filling material for dikes, roads and artificial islands.

The consolidation of slurries is a complex process that depends on many physico-chemical aspects. Sediment particles in suspension are interacting and this interaction can lead to time-dependent flocculation and settling rates. Settled flocs will create a very open and fluffy bed that will consolidate over time, exhibiting large strains during the primary consolidation regime. At the same time, a substantial amount of fine particles can remain (unflocculated) in suspension due to Brownian motion.

The aim of our research is to link the suspension phase to the consolidating phase in a single model. This model combines a traditional 1D large-strain consolidation model with 1D advection-diffusion model as derived in colloid science in the suspension phase. The model is subsequently tested on various samples from the aforementioned projects.

RÉSUMÉ: De nombreux projets (Marker Wadden, Kleirijperij) ont récemment été lancés aux Pays-Bas, qui incluent la réutilisation de matériaux de dragage provenant de chenaux envasés. Ces matériaux de dragage sont déposés dans de grands bassins et laissés à consolider. Lorsque le drainage et le développement de la résistance du matériau sont jugés suffisants, celui-ci est utilisé comme matériau de remplissage pour les digues, les routes et les îles artificielles.

La consolidation des vases est un processus complexe qui dépend de nombreux aspects physico-chimiques. Les sédiments en suspension interagissent et cette interaction peut donner lieu à des taux de flocculation et de sédimentation dépendants du temps. Les agrégats sédimentés créeront d'abord un lit très ouvert et floconneux qui se consolidera au cours du temps, présentant de larges déformations pendant le régime de consolidation primaire. En même temps, une quantité importante de particules fines (non flocculées) peut rester en suspension par l'action du mouvement Brownien.

Le but de notre recherche est de lier la phase de suspension à la phase de consolidation en un modèle unique. Ce modèle combine un modèle traditionnel de consolidation 1D à grandes déformations avec un modèle d'advection-diffusion 1D telle qu'il est dérivé en science des colloïdes pour la phase de suspension. Le modèle est ensuite testé sur divers échantillons provenant des projets susmentionnés.

Keywords: consolidation; colloid; slurries; advection-diffusion

1 INTRODUCTION

The topic of dewatering and consolidation of soft slurries has regained interest over the past years as natural slurries (such as dredged material from silted channels) are thought to be economically good alternatives to the use of sand for building or filling materials. Large-scale projects have been initiated in the Netherlands (Marker Wadden, Kleirijperij) to study the feasibility of large-scale dewatering and strengthening of soft slurries.

Soft slurries are a mixture of silts and clays, in the presence or not of organic matter, in a water medium. Two main phases are distinguished once the material is disposed into a dewatering pond: a settling phase, where the particles can be in suspension or in a gel state (interconnected particles) but where effective stresses are negligible and a consolidation phase, where the particles are in a gel state and where effective stresses between particles play a role.

Depending on the composition of the slurries, in particular the sediment particle size and the presence of organic matter, different settling and consolidation behaviours can be observed. The behaviour of the smallest particle size class ($< 50 \mu\text{m}$) is governed by colloidal forces, i.e. interaction between particles which can lead to aggregation (flocculation) or repulsion between particles (Russel 1991, Dhont 1996). Studying the settling phase of a flocculating suspension is very challenging, as particles undergo changes in velocities (accelerations) over time as their sizes increase. In most settling studies in the laboratory it is assumed that flocs have reached their equilibrium size prior settling. This equilibrium size is in the range $[10 - 1000 \mu\text{m}]$ and heavily depends on environmental conditions (water composition, presence of organic matter, hydrodynamics) and particle composition (size, shape, surface charge).

The key parameter for modelling the settling of flocs is their density, which is, as is their size and shape, depending on the floc composition and growth history (Mietta 2009).

From various experimental studies it has been demonstrated that on average the density of a floc is an exponentially decreasing function of its size (Dyer 1999), which could be well interpreted using a fractal approach. The concept of fractal has subsequently been introduced to describe the settling and consolidation of flocs (Merckelbach 2000).

Depending on their composition, and in particular their organic material content, natural soft slurries can be composed of either flocculated and non-flocculated material (see Fig.1) or a combination thereof.

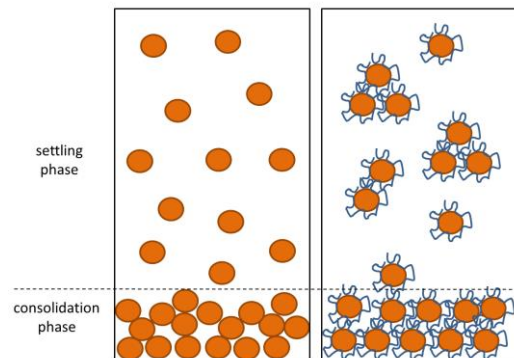


Figure 1. Schematic representation of an unflocculated (left) and flocculated (right) system. In the flocculated state, aggregates (flocs) are formed thanks to the presence of organic material (in blue)

Settling models for both flocculated and non-flocculated clays will be discussed. The settling and consolidation phase of natural slurries of the Markermeer lake (The Netherlands) will be studied, using a combination of settling and consolidation models.

2 SETTLING PHASE

2.1 Settling of unflocculated clay

The simplest case to be studied is the one involving dilute suspensions of fine particles, in a laminar flow. In that case, Stokes settling velocity can describe the change in water / suspended particles interface over time. For non-dilute suspensions, corrections have been introduced to account for the fact that the settling is hindered by the presence of other particles (Richardson 1954):

$$h = h_0 - v_0 \cdot (1 - \phi_s / \phi_{gel})^m \cdot t \quad (1)$$

where $h(m)$ is the position of the water / suspension, $h_0(m)$ the initial height, $v_0(m/s)$ is Stokes settling velocity, $\phi_s(-)$ is the solid volume fraction, $\phi_{gel}(-)$ is the volume fraction at gelling (i.e. when particles are forming an interconnected network), $t(s)$ is the time and m is an empirical coefficient. Studies have demonstrated that for colloidal particles, m varies usually between 3 and 6 (Dhont 1996).

An example is given in Figure 2, where it can be observed that the effect of hindered settling is negligible, since the clay concentration is very low. We used for ϕ_{gel} the value for random close packing of hard spheres as an estimate. The experiment has been performed using a fine-grained clay, devoid of organic matter. Due to the size of the particles ($10 \mu m$) one finds that the Reynolds number is low and therefore the flow is laminar. For larger particles in the non-laminar regime, additional corrections have been introduced to the settling velocity expression (Camenen 2011).

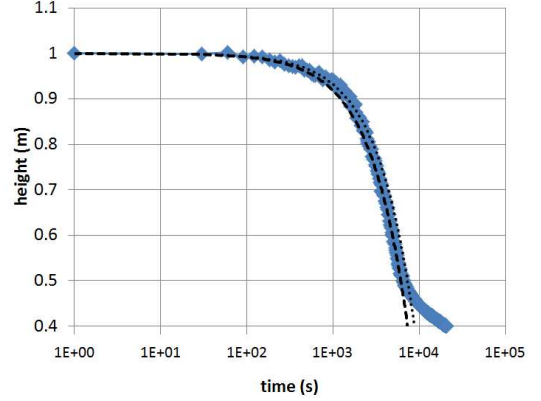


Figure 2. Water/ suspended clay interface as a function of time. Clay concentration is 50 g/L and the mean particle size is $10 \mu m$. Dashed line: settling profile according to Stokes ($m=0$ in eq.(1)), full line: profile found using eq.(1) with $m=5$ and $\phi_{gel} = 0.74$. Data is taken from (Ibanez 2018)

For the same type but smaller-sized particles, the interface between water and suspension becomes fuzzy, as Brownian diffusion comes into play. This diffusive term, related to the osmotic pressure in the suspension, plays the same role as the effective stress in the Gibson consolidation equation (Toorman 1996).

This is illustrated in Figure 2. To generate the graphs the following advection-diffusion equation was solved to find $\phi_s(z, t)$:

$$\begin{aligned} \frac{\partial \phi_s}{\partial t} - v_0 \frac{\partial}{\partial z} [\phi_s f(\phi_s)] \\ = D_0 \frac{\partial}{\partial z} \left[f(\phi_s) \frac{\partial [\phi_s Z(\phi_s)]}{\partial z} \right] \quad (2) \end{aligned}$$

where $f(\phi_s) = (1 - \phi_s / \phi_{gel})^m$ is associated to the advective term, $D_0(m^2/s)$ is the diffusion coefficient of the particle and $Z(\phi_s)$ is the Carnahan-Starling equation of state (Dhont 1996) and is associated to the diffusive term.

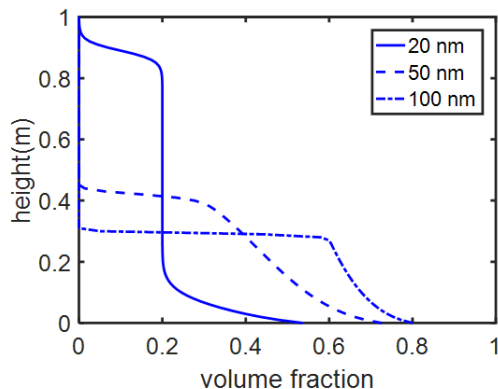


Figure 3. Theoretical water/suspended clay interface for different clay particle sizes after 1000 days. Initial clay concentration is 500 g/L ($\phi_s = 0.2$).

One can observe the fuzziness of the interface for the 20 nm particles as the volume fraction varies gradually close to the interface. Diffusion is dominating the system and the 20 nm particles have barely settled. Another cause for the diffusive term can be the electric repulsion between particles, which also prevent them from aggregating. This changes the expression for the osmotic pressure and hence $Z(\phi_s)$ (Bowen 1996, Carriere 2007). The profiles obtained remain however very similar to the ones presented in Fig.3.

In natural systems very fine particles usually always remain in suspension, as the water hydrodynamics will ensure that settling is not possible and the long-term profiles given in Fig.3 are not observed: the concentration of fines will remain constant over the whole water column.

2.2 Settling of flocculated clay

Natural slurries are usually containing a significant amount of organic material, such as extracellular polymeric substances (EPS) and dissolved organic carbon (DOM) which can induce flocculation between the mineral sediment particles. In that case, after a transient time, an equilibrium floc size will be reached which depends on sediment and water

composition as well as on local hydrodynamic conditions. In settling ponds, which is our topic of interest, these conditions are energetically low, and the flocs are assumed to have reached their equilibrium size prior their deposition in the ponds.

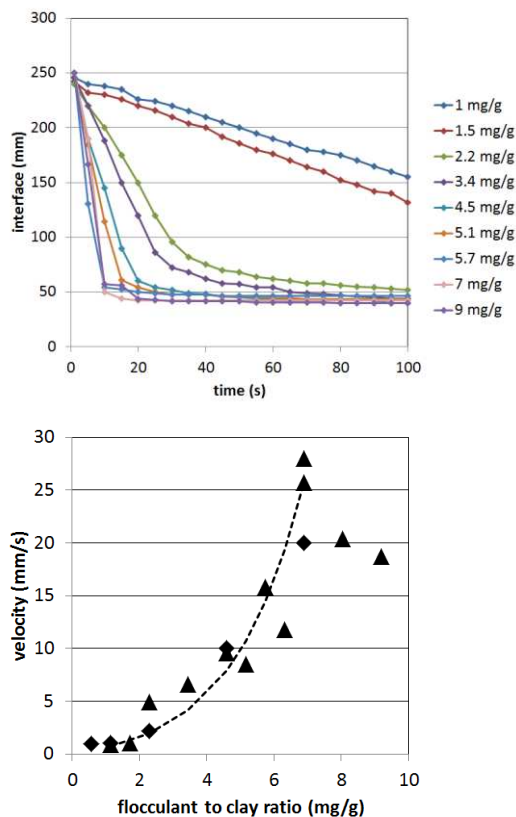


Figure 4. Upper panel: water/settling clay interface for a flocculated clay. Original size of the clay particles is 10 μm and clay concentration is 0.7 g/L. The different EPS (flocculant) to clay ratios are indicated in the legend. Bottom panel: estimated settling velocities from the data presented in the upper panel. Data is taken from (Ibanez 2018)

The type and amount of organic matter present in the system is determinant for the settling. Experiments were performed in the laboratory, where different amounts of synthetic EPS (Zetag 7587, a cationic polyelectrolyte from BASF) were added to clay suspensions. The settling velocity of the formed flocs was then estimated

from the water/slurry interface as function of time, see Fig. 4.

At low EPS to clay ratio, the settling phase can be predicted using Stokes ($m=0$ in eq.(1)) as was shown in Fig.1. At higher ratios the settling velocity is increased, which is linked to the fact that the particle size is increased due to flocculation (see Fig.5).

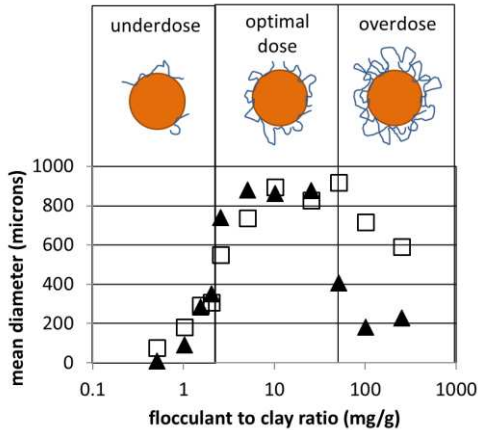


Figure 5. Mean particle size as function of time and flocculant concentration. Full symbols: after 120 s; open symbols: after 600 s. Data is taken from (Ibanez 2018)

At the optimal ratio (about 5 mg/g) the settling velocity reaches a maximum, and it decreases for larger ratios. These larger ratios correspond to an overdose concentration of flocculant: there is so much EPS in suspension that the extension of the polymeric hairs slow down the flocculation process (see Fig.5) and also hinder the settling (see Fig.4).

As the floc size is increasing with EPS to clay ratio, its density is decreasing with size, as illustrated in Fig.6. The data has been obtained by analysing the settling velocities of thousands of flocs recorded by video microscopy. The effective density ($\rho_{floc} - \rho_w$) has then been estimated using Stokes. The fit is obtained by adjusting the fractal dimension in eq.(3).

$$(\rho_{floc} - \rho_w) = (\rho_s - \rho_w) \cdot \left(\frac{R}{R_{floc}} \right)^{3-D} \quad (3)$$

where ρ_{floc} , ρ_w and ρ_s are the density (g/L) of a floc, water and solid, R is the size of a clay particle (10 μm) and R_{floc} the size of a floc (μm).

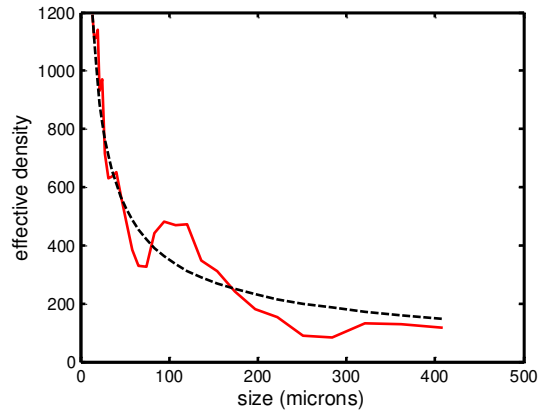


Figure 6. Effective density of flocs as function of their size. A fractal dimension of $D= 2.39$ was found using eq.(3).

3 CONSOLIDATION

The transition between settling phase and consolidation phase occurs at the gel point (Buscall 1990) and on Fig.2 is represented by the point where the theoretical fit deviates from the experimental data. At that point a sharp change in slope is observed which corresponds to the water/suspension interface meeting the suspension/bed interface. As the bed is consolidating with a much smaller velocity than the settling particles, this leads to the change in slope. When the volume fraction of suspending particles is high, it happens that from the start the particles (flocs) are forming an interconnected network and the transition between the interfaces is smooth, as shown in Fig.7.

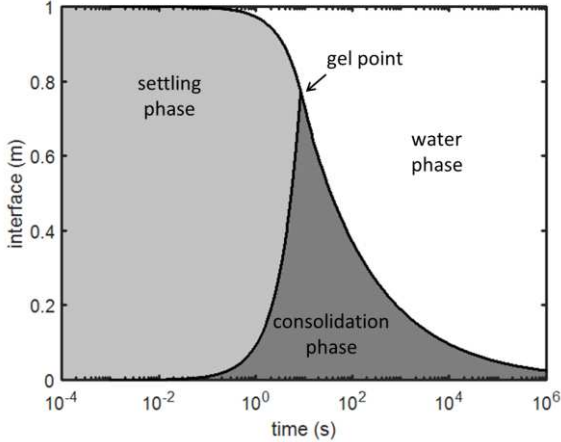


Figure 7. Theoretical evolution of the water/settling and the settling/bed interfaces as function of time for an interconnected network of flocs. In the settling phase the volume fraction at any height is given by the initial volume fraction.

In the case of very small unflocculated particles, as shown in Fig.3, a physical contact between particles might never be reached as Brownian motion will dominate the system. There will be a density profile, more or less pronounced, depending on the particle size, but no effective strength will be developed.

In the case of flocculated material where all flocs are interconnected, the settling phase as represented in Fig.7 corresponds to the collapse of the gelled structure under its own weight. As the flocculated material is assumed to be homogeneous at start, the volume fraction in the settling phase is constant. The flocs have a very open structure, reflected in their fractal dimension (see Fig.6), and can contain up to 90% of water. At collapse the flocs will release their water as they are compacted, and this process starts at the bottom of the water column (consolidation phase). This can be measured by recording the excess pore water pressure (Been and Sills 1981). This part of the consolidation is referred to as primary consolidation. When the excess pore water pressure has been dissipated, a slow consolidation regime (secondary consolidation) is found, due to the compression

of the solid skeleton under its own weight (creep).

In 2000, a new formulation for describing the consolidation of soft slurries was presented and validated for different natural muds from Ems-Dollard Estuary and the Caland-Beer Channel (the entrance channel of the Port of Rotterdam), in the Netherlands (Merkelbach 2000). This model is based on an advection-diffusion equation quite similar to eq.(2), but where the advective term is now expressed as a permeability and the diffusive term as an effective stress. Both permeability K (m/s) and effective stress σ_{sk} (Pa) are functions of the same fractal dimension:

$$K(\phi_s) = K_k[\phi_s]^{-n} \quad (4)$$

$$\sigma_{sk}(\phi_s) = K_\sigma[\phi_s]^n \quad (5)$$

where $n = 2/(3 - D)$ and D is the fractal dimension (a number between 2 and 3). The advection-diffusion can be solved analytically for the secondary consolidation phase and the end volume fraction profile ϕ_∞ is given by

$$\phi_\infty(z) \approx \phi_\infty(z=0) \left[1 - \frac{z}{h_\infty} \right]^{\frac{1}{n-1}} \quad (6)$$

where

$$\phi_\infty(z=0) \approx \left[\frac{\phi_0 H (\rho_s - \rho_w) g}{K_\sigma} \right]^{\frac{1}{n}} \quad (7)$$

and

$$h_\infty \approx \frac{n}{(n-1)} \frac{\phi_0}{\phi_\infty(z=0)} H \quad (8)$$

where H (m) and $\phi_0(-)$ are the initial height and volume fraction (the column has a uniform volume fraction at start) and h_∞ (m) is the final height on the bed.

From fitting the density profile at the end of consolidation, the parameters K_σ and D can be obtained. This is illustrated in Fig.(8)

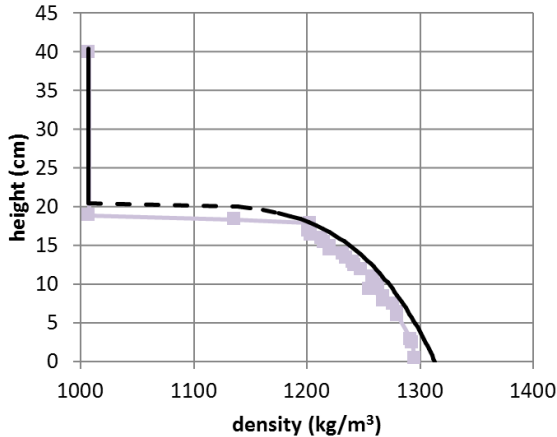


Figure 8. Final density profile of a flocculated bed of concentration 200 g/L ($\phi_s = 0.077$). Symbols: measurements by Ultra-sonic High Concentration Meter (UHCM). Black line: fit using eqs.(5-8), with $K_\sigma = 6.5 \cdot 10^6$ Pa and $D = 2.65$. Data from (van den Bosch 2016). The sediment is from lake Markermeer.

The parameter $K_k = 2.0 \cdot 10^{-12}$ m/s is then found by fitting the time evolution of the interface, keeping K_σ and D constant. This was done for the time evolution of the 200 g/L sample. As can be seen in Fig.9, the model is then able, using the same parameters, to predict the time evolution of samples of higher concentrations. For lower concentrations, the flocs are not interconnected at start (reflected in the change in slope around 10^4 s for the 100 g/L sample). The settling phase can then be fitted (dashed lines) using eq.(1), and we found $\phi_{gel} = 0.075$ (195 g/L), which agrees with the fact that the 200 g/L sample is in a gel state. We used $v_0 = 1.5$ mm/s which is the Stokes settling velocity for 40 μ m particles, in agreement with the fact that the clay was sieved through a 63 μ m sieve prior dispersion. Using the same ϕ_{gel} and v_0 the settling phase of all samples below gelling could reasonably be predicted. The final consolidation of each of these samples could also be predicted using the same K_σ and D values found from the fit of the 200 g/L sample. For the lowest clay

concentration (40 g/L) there is a good transition between the settling and the consolidation model. This is less the case for the 100 g/L sample. In order to improve the fit, it was necessary to double the value of K_k (red line). The reason of the mismatch could be linked to the limitation of the model in the suspension/gel transition range (just below 200 g/L as discussed above) or due to the fact that the 100 g/L sample was not well-mixed at the onset of the experiment and that a sediment layer was already present at the bottom of the column. To check this last hypothesis, one should then know the density profile at start (not known for the present set of experiments).

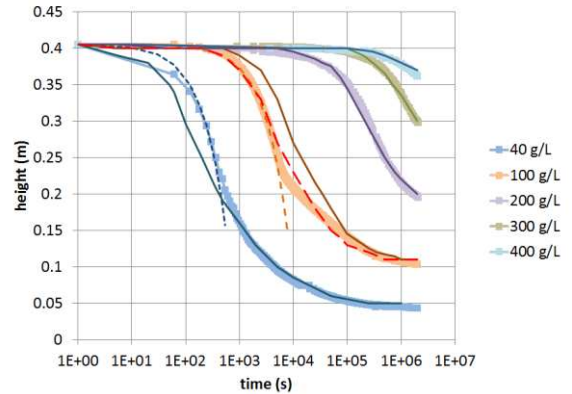


Figure 9. Time evolution of water/sediment interface for different sediment concentrations. Symbols are measurements. Lines are fits (see text). Full lines: $K_\sigma = 6.5 \cdot 10^6$ Pa, $K_k = 2.0 \cdot 10^{-12}$ m/s and $D = 2.65$. Data from (van den Bosch 2016). The sediment is from lake Markermeer.

4 CONCLUSIONS

The settling of (un)flocculated suspensions can be understood and modelled, thanks to the work done in colloid science on this topic.

The model available for the consolidation of soft slurries was tested and it was found that it could reproduce the time evolution of natural slurries above gelling ($\phi_0 > \phi_{gel}$). A discrepancy between model and experiments

were observed for $\phi_0 < \phi_{gel}$ for one of the sample. Nonetheless all final bed heights for all clay concentrations (40 – 400 g/L) could be predicted with the same parameters that were found from fitting the 200 g/L density end profile. The model is currently tested for a large variety of samples, as function of sample composition.

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