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SEDIMENTATION OF CONCENTRATED SUSPENSIONS IN NON-NEWTONIAN FLUIDS

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ABSTRACT. The behavior of polydisperse concentrated solid suspensions (ε smaller than 0.95) of quartz sand in carboxymethylcellulose (CMC) aqueous solutions (with 0.5%, 1.0% and 1.5%, in weight) during the sedimentation process was investigated. The influence of solid size, solid concentration and rheological parameters of CMC disperse fluid with a non-Newtonian behavior on the settling kinetics has been theoretically accounted. The experimental hindered settling velocities determined graphically by Kynch method and those expressed by Richardson-Zaki equation are compared and analyzed.

Keywords: *particle settling velocity, particle size distribution, hindered settling velocity, Richardson-Zaki equation, rheological parameters.*

INTRODUCTION

The sedimentation of particles in non-Newtonian fluids is an essential problem in the case of suspensions storage for a long time, in many pharmaceutical products, paints, detergents, agro-chemicals, emulsions and foams being often desirable to keep the active component uniformly suspended. The stability of these systems can be quantitatively determined by the settling velocity of suspended particles.

The settling velocity of a single particle in a viscous fluid, under steady-state conditions, in a large vessel is easily estimated by balancing the weight of the particle with the buoyancy and drag forces, as Stokes' equation (1) shows:

$$w_s = \frac{g \cdot d^2(\rho_s - \rho)}{18 \cdot \eta} \tag{1}$$

where: *d* is the average particle size (m), ρ_s – the solid density (kg/m^3), ρ - the disperse fluid density (kg/m^3), η - the fluid viscosity (Pa·s), *g* – the gravitational acceleration (m/s^2).

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Experience with Newtonian fluids has shown that the hydrodynamics of systems consisting of single particles, drops or bubbles serves as a useful starting point for understanding the mechanism of the more complex multiparticle systems [1]. Determination of the settling velocity in polydisperse concentrated suspensions, which are far more common, named hindered settling velocity, is a difficult problem due to the complexity of the particle-particle and particle-fluid interactions [2, 3].

The expression proposed by Richardson-Zaki for Newtonian fluids, equation (2), applied at values of terminal Reynolds number Re_t up to about 2 [4], is the most popular empirical equation used in modeling and numerical simulation of hindered settling velocities [5-8]:

$$w(c) = w_S(1 - C_v)^z = w_S \cdot \varepsilon^z \qquad (2)$$

where: *w(c)* is the hindered settling velocity (m/s), w_S - the Stokes' velocity (m/s), which graphically represents the extrapolation of the velocity to the voidage equal 1, related to a single particle terminal settling, C_v - the solid volume concentration (m^3/m^3), ε - the voidage or void fraction (-) and *z* – the sedimentation exponent (-).

Parameter *z* was found to be a function of the flow regime, expressed by the terminal Reynolds number Re_t, equation (3), and the particle to column diameter ratio *d/D* (Table 1) [4, 9]. Smaller vessel dimensions lead to a reduction of the settling velocity, more pronounced as the particle diameter, *d*, becomes comparable to the vessel diameter, *D*.

Table 1. Values of the parameter z as recommended by Richardson-Zaki.

$Re_t < 0.2$	$z = 4.65 + 19.59(d/D)$
$0.2 < Re_t < 1$	$z = 4.35 + 17.5(d/D)\,\mathrm{Re}_t^{-0.03}$
$1 < Re_t < 200$	$z = 4.65 + 18(d/D)\,\mathrm{Re}_t^{-0.1}$
$200 < Re_t < 500$	$z = 4.45\,\mathrm{Re}_t^{-0.1}$
$Re_t > 500$	$z = 2.39$

The terminal Reynolds number corrected for power-law liquids has the expression [1]:

$$\mathrm{Re}_t = \frac{\rho \cdot w_S^{\,2-n} \cdot d^n}{k} \qquad (3)$$

where: w_S is the Stokes' velocity corrected for the sedimentation in non-Newtonian fluids (m/s), d - the average particle size (m), ρ - the disperse fluid density (kg/m^3), k – the fluid consistency coefficient (Pa·s^n), n – the flow behavior index (-).

In creeping regime ($Re_t < 2$), the settling velocity of a spherical particle in non-Newtonian fluids following power-law can be estimated by equation (4) [1]:

$$w_s = \frac{g \cdot d^{n+1}(\rho_s - \rho)^{1/n}}{18 \cdot k \cdot X(n)} \tag{4}$$

where: ρ_S is the disperse fluid density (kg/m^3), *X(n)* - the deviation factor, a function of flow index *n* [1, 10].

For $Re_t > 2$, the parameter *z* is a function of Archimedes number and (*d/D*) ratio, and is given by equation (5) [1]:

$$\frac{4{,}8 - z}{z - 2{,}4} = 0.0365 Ar^{0{,}57}\left[1 - 2{,}4(d/D)^{0{,}27}\right] \tag{5}$$

where for power-law liquids, the Archimedes number, *Ar*, is defined by equation (6):

$$Ar = \xi \operatorname{Re}^{2/(2-n)} = \frac{4}{3} g d^{(2+n)/(2-n)}(\rho_s - \rho)\rho^{n/(2-n)} k^{2/(n-2)} \tag{6}$$

It is obvious that the settling velocity shows in the case of non-Newtonian fluid a stronger dependence on particle diameter and density difference than in a Newtonian fluid.

The sedimentation theory by Kynch was used as graphical approach in our study in order to find the settling velocity of quartz sand suspension over time, keeping in mind, that in this method the wall effects are neglected and uniform particle shape and size are assumed [11]. The Kynch method involves constructing tangents to the settling curves, and the slope of each of these tangents is the velocity at that time.

The aim of the present study was to compare the theoretical settling velocities predicted by Richardson-Zaki equation corrected for non-Newtonian fluids with those experimentally determined by Kynch method, in the case of sedimentation of quartz sand in carboxymethylcellulose (CMC) solutions with different concentrations. The influence of particle size determined by particle size distribution (PSD), quartz sand concentration and rheological parameters of disperse fluid (CMC) on theoretical settling velocities are accounted and analyzed.

RESULTS AND DISCUSSION

Rheological characterization of CMC solutions

The flow curves of carboxymethylcellulose (CMC) solutions, based on the dependence of the shear stresses on the shear rates, shown in Figure 1, and the diagram of apparent viscosities versus shear rates, shown in Figure 2, indicate a typical shear-thinning or pseudoplastic behavior [12, 13].

The decrease of apparent viscosity (η_{app}) with shear rate ($\dot{\gamma}$) follows an Ostwald power-law model, described by equation (7):

$$\eta_{app} = k\dot{\gamma}^{n-1} \quad (7)$$

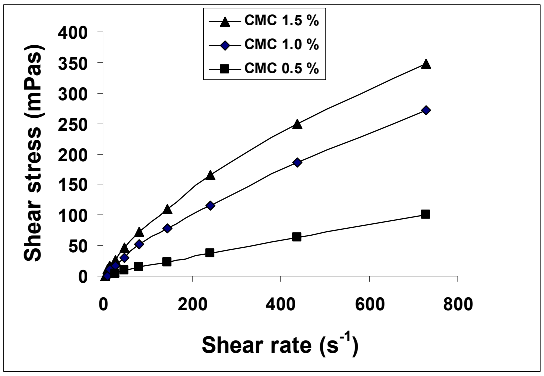

Figure 1. Flow curves of CMC solutions.

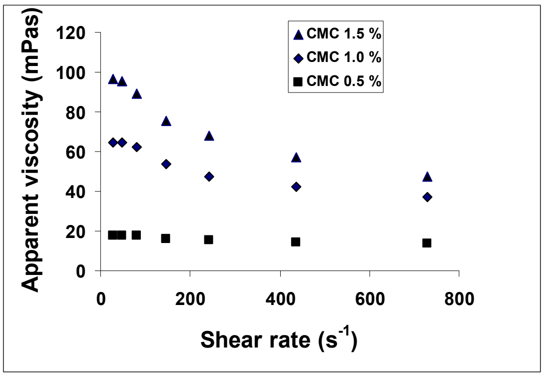

Figure 2. Apparent viscosity vs. shear rate.

The interpretation of experimental data leads to the average value of consistency coefficient *k* and the flow behavior index *n* shown in Table 2.

Table 2. Values of *k* and *n* at different concentration of CMC.

CMC concentration (%)	*k* (mPa·s)	*n*
1.5	287.82	0.7846
1.0	115.69	0.8471
0.5	26.705	0.9050

Sedimentation study

Experimental settling velocities

Typical settling curves of quarz sand (d < 200 μm) suspensions (C_v ranging from 0.088 to 0.265) in 0.5% CMC solution are shown in Figure 3.

The incipient settling velocities are determined from the slope of linear portion of each settling curve. Results clearly show that the settling velocity decreases with the increase of solid particle concentration.

Similar interpretation are done for the sedimentation of quarz sand suspensions in 1.0% and 1.5% CMC solutions.

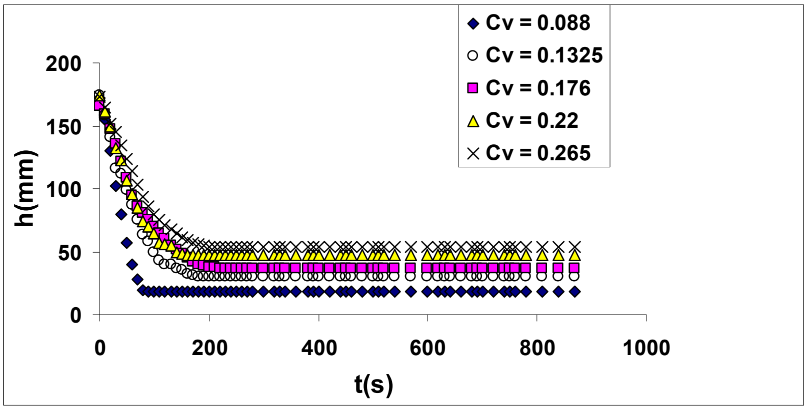

Figure 3. Sedimentation curves of quartz sand in 0.5% CMC solution.

Theoretical settling velocities

The theoretical Stokes' velocities are calculated by equation (4) with the obtained values of rheological parameters *k* and *n* and the average particle size considered one of the follows (Table 3) [13]:

a. d_{mode} - the equivalent diameter corresponding to the top (maximum) of the differential particle size distribution (PSD) curve (Figure 4);

b. d_{50} - the average equivalent diameter considering the cumulative PSD curve at T% = R% = 50% (Figure 4);

c. d_m - the average weighted diameter of particles by equation (8):

$$d_m = \frac{d_{m1}p_1 + d_{m2}p_2 + ... + d_{mn}p_n}{p_1 + p_2 + ... + p_n} = \frac{\sum_{i=1}^{n} d_{mi}p_i}{100} \quad (8)$$

where: d_{mi} is the arithmetic mean of two consecutive mesh sieve of sieve shaker used in PSD analyze, and p_i – the percentage of each retained fraction.

The theoretical hindered settling velocities are estimated by Richardson-Zaki equation, all terminal Reynolds numbers verified by equation (3) being lower than 2.

No significant influence of *d/D* ratio was observed because the solid particles are much smaller (*d* < 200 μm) than the cylinder diameter (*D* = 28 mm), and so the wall effect can be ignored.

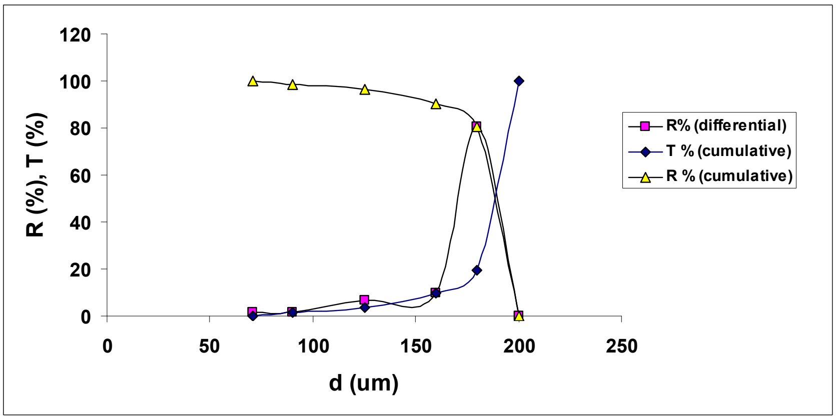

Figure 4. Particle size distribution (PSD) curves.

Table 3. Average particle size.

Particle fraction	d_{mode} (μm)	d_{50} (μm)	d_m (μm)
< 200 (μm)	180	190	180

Experimental settling velocities and corresponding Richardson-Zaki predictions as function of voidage in tested CMC solutions are shown in Figure 5 (a, b, c).

It is obvious that there are considerable differences between theoretical settling velocities estimated by Richardson-Zaki equation, w_t, and experimental values graphically determined by Kynch method, w_{exp}. In all analyzed cases, the theoretical velocities are larger than those experimentally determined.

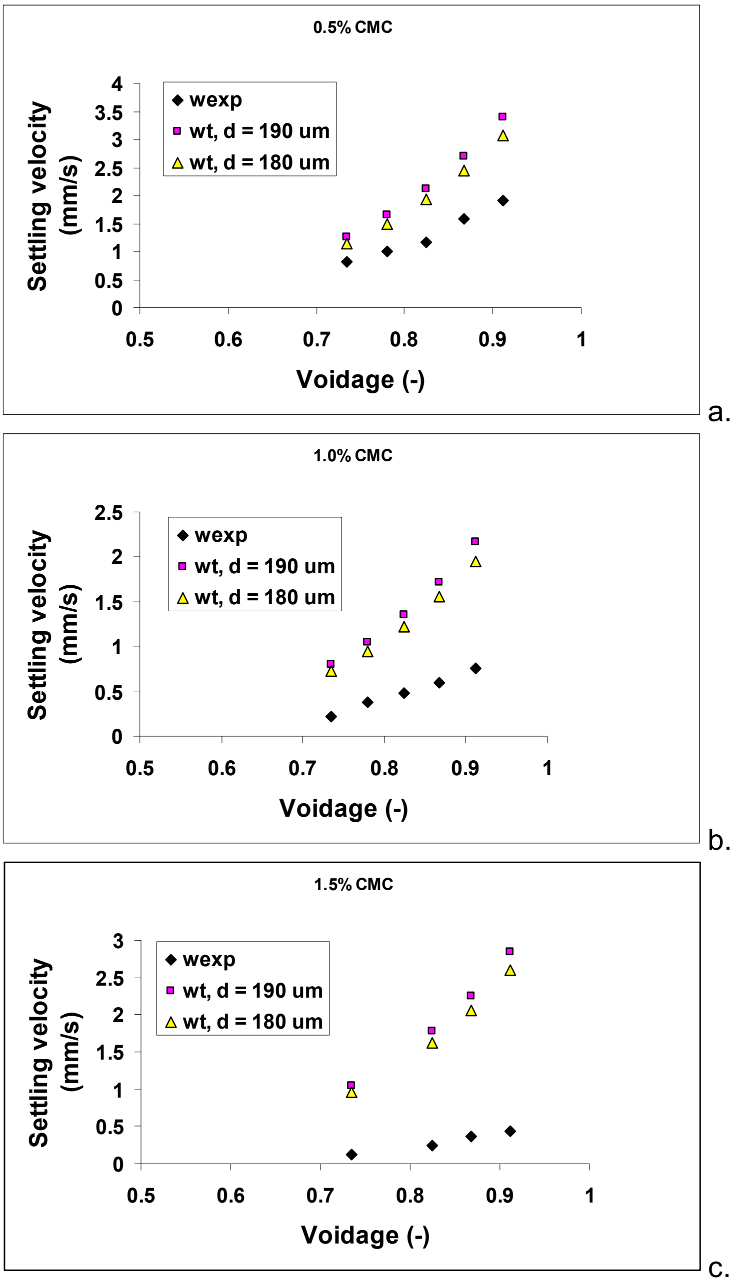

Figure 5. Experimental and theoretical settling velocities vs. voidage.

For the 180 μm average particle size, the ratio w_t/w_{exp} is about 1.5 - 1.6, in the case of sedimentation in 0.5% CMC solution, 2.5 – 3.3 in 1.0% and 6.0 – 7.5 in 1.5% CMC solution. This means that the difference between theoretical and experimental velocities decreases with the decrease of CMC concentration.

In the same time the difference between theoretical and experimental velocities is influenced by the solid concentration. Generally, the difference decreases slowly with the increase of quartz sand concentration in the same CMC solution.

In order to determine the values of sedimentation exponent *z*, the experimental settling velocity w_{exp} and voidage are plotted on log-log coordinates, as a linearized Richardson-Zaki equation (Figure 6).

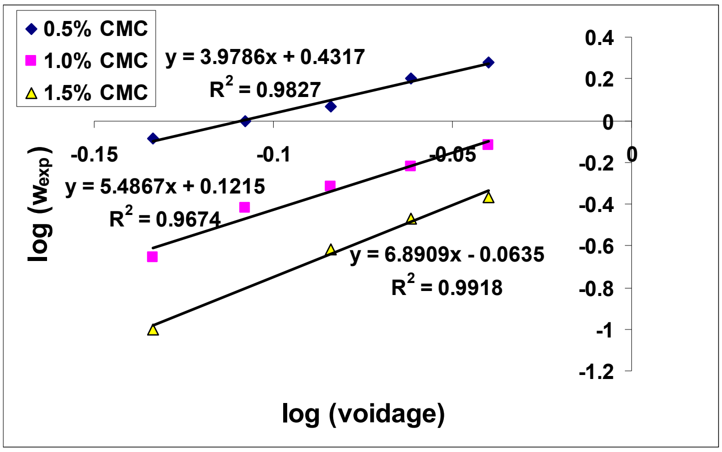

Figure 6. Log-log plot of experimental settling velocity and voidage.

The obtained values of *z* parameter are shown in Table 4.

Table 4. Values of sedimentation exponent.

0.5% CMC	1.0% CMC	1.5% CMC
$z \cong 4.0$	$z \cong 5.5$	$z \cong 6.9$

The obtained value of sedimentation exponent *z* is lower than Richardson-Zaki value (z=4.65) in solution of 0.5% CMC, and larger than Richardson-Zaki value in 1.0% and 1.5% CMC solutions, showing an increase with the increase of CMC concentration. Excepting the sedimentation exponent *z* obtained in 1.5% CMC solution, the other two values are comparable with those founded in literature [14, 15].

It is possible that the hindered settling effect of quartz sand sedimentation in CMC solutions with non-Newtonian behavior, especially in 1.0% and 1.5% concentration, to be greater than could be estimated by theoretical relations. Other secondary effects (e.g. adsorption of CMC on quartz sand) which can induce a higher friction between particles are not excluded.

CONCLUSIONS

The Stokes' velocities and the theoretical velocities estimated by Richardson-Zaki equation (z=4.65) were calculated, using experimental values of rheological parameters n and k for each CMC solution and the average particle size of quartz sand.

The obtained values were compared with experimental values determined by Kynch method. Generally, the theoretical velocities values predicted by Richardson-Zaki expression were larger than those experimentally obtained, the difference between theoretical and experimental velocities decreasing with the decrease of CMC concentration and with the increase of solid concentration.

The sedimentation exponent z determined by experimental data was different from the Richardson-Zaki value (z=4.65), comparable with the value founded in literature in CMC solution of 0.5% and 1.0%. It is possible that the hindered settling effect in non-Newtonian fluids to be greater than could be estimated by theoretical relations.

For the important practical cases of concentrated solid suspensions in non-Newtonian fluid it is indicated to evaluate the settling velocities by experimental measurements.

EXPERIMENTAL SECTION

Rheological measurements of CMC solutions of different concentration (0.5%, 1.0% and 1.5%, in weight) have been carried out at room temperature 20 ± 1^0C, using the rheometer Rheotest-2, and the system S/S1. Three replicates were performed for each new solution. The fluid rheological characteristics are shown in Table 2.

Experimental settling velocities were determined by the sedimentation curves considering the Kynch method. Experimental runs were carried out in cylinders with internal diameter of 28 mm and 300 mm tall.

The quartz sand particles (d < 200μm) were sieved in order to determine the average particle size using a sieve shaker with sieve mesh of 200, 180, 160, 125, 90 and 71 μm.

Different suspensions were made by mixing quartz sand with CMC solutions (0.5%, 1.0% and 1.5%, in weight). Prior each settling experiment, the blends were strongly mixed by shaking, directly in the cylinder were the tests are done, in order to obtain a well dispersed initial state. After mixing, the cylinder was allowed to stand, and the height of quartz sand was noted at regular intervals until no further sedimentation occurs. The boundary between the settling suspension and the supernatant phase were determined visually for each sample, as a function of time in order to establish the sedimentation curves. Each experiment was performed in triplicates.

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