Technical Sciences
Academy of Romania
www.jesi.astr.ro

Journal of Engineering Sciences and Innovation
Volume 9, Issue 3 / 2024, p. 277 - 286

C. Chemical Engineering, Materials Science and Engineering

Received ***21 Apryl 2024*** *Accepted* ***3 September 2024***
Received in revised form ***23 August 2024***

Problems of Stokes' law application in determining the settling velocity of clays

NAOMI A. OGOLO[1*], MIKE O. ONYEKONWU[1], ABBEY T. MICHAEL[2]

[1]*Institute of Petroleum Studies, University of Port Harcourt, Rivers State, Nigeria*
[2]*Physics Department, Faculty of Science, University of Port Harcourt, Rivers State, Nigeria*

Abstract. Clay sedimentation is important for various applications including settling of drilling mud during downtime in drilling operations, and it is essential to model such processes. A widely accepted theory that explains the kinetics of dispersed particles under gravitational pull in a quiescent medium is Stokes' law. This law is commendable and reliable for modelling the settling velocity of particles but has been reported to be inadequate for modelling the settling velocity of clays. This has prompted a re-examination of the law with regard to assumptions made in deriving it, and it was found that several factors associated with clay sedimentation were not considered in the law. These factors include particle shape, size, interactions, flocculation, salinity, pH value, concentration and hindrance velocity of settling. Hence, this paper discusses these factors and how they defy assumptions made in Stokes' law, thereby rendering the law unsuitable for modeling clay settlement in liquids.

Key words: particles, flocculation, shape, salinity, settling velocity, concentration.

1. Introduction

Clay minerals are very important raw materials and the primary types are Kaolinite, Montmorillonite and Illite, used in various industries such as in the construction, ceramic and pharmaceutical industries. Geologists, agricultural, civil, environmental, and petroleum engineers are all interested in studying clay mineral characteristics because they are unavoidable during field operations. In the petroleum industry, clays are encountered in the formation during oil and gas production and sometimes they constitute a nuisance. Cap rocks that seal hydrocarbon reservoirs are partly composed of clays. In drilling of oil and gas

*Correspondence address: amoniogolo@yahoo.com

wells, Montmorillonite also called Bentonite is used in preparing drilling fluids that perform several functions including prevention of kicks and blowouts which disrupts drilling operations and pollutes the environment. In fact, during downtime in drilling operations, drilling muds have the tendency of settling at the bottom of the well, necessitating that the particle settling velocity of Montmorillonite be studied. Such settlements are problematic because it undermines the functions of drilling fluids; hence additives that promote clay stability are desired. Studying the effectiveness of such additives also requires particle settling velocity studies. It is therefore crucial to examines models that have been used to predict particle sedimentation in vertical columns in order to ensure adequacy, high confidence level and reliability. In this work however, the focus is on Stokes' law because it is fundamental in determining the falling rate of particles under gravity in a vertical column.

Stokes' law of particle settlement under gravity is popular and it is used to describe the settling velocity of particles in a column of fluid. The law states that the force that retards a sphere moving through a viscous fluid is directly proportional to the velocity, radius of the sphere and viscosity of the fluid. This law is reliable for particle sizes smaller than approximately 0.1 mm and it is mathematically expressed in Eq. 1 as:

$$V_s = \frac{2r^2 g(\rho_p - \rho_f)}{9\mu} \qquad \text{Eq. 1}$$

where

V_s = Velocity of settling (m/s)

ρ_p = Mass density of the particles (kg/m^3)

ρ_f = Mass density of the fluid (kg/m^3)

g = Acceleration due to gravity (9.81m/s^2)

r = Radius of the particle (m)

μ = Viscosity of the Fluid (kg/(ms))

1.1. Assumptions of Stokes' law and implications

Stokes' law and its application to particle settlement in a medium were formulated based on certain basic assumptions. These assumptions and their implications are outlined below:

1. Particles are rigid, smooth and spherical, and must be of uniform density (or constant). Ordinarily, the particle density is considered to be between 2.65 to 2.6g/cm^3 (but could vary between 2.0 to 3.2g/cm^3).
2. Particles must be sufficiently large (greater than 0.001mm) to avoid Brownian motion. Particles less than 0.0002m exhibit Brownian motion and this adversely affects the settling velocity.
3. There should be no interaction between individual particles in the medium; falling particles are not hindered or affected by walls of the vessels or other falling

particles. This means that particles must not interfere with each other during the period of fall.

4. Temperature and viscosity of the water should be constant throughout the settling period. This is because settling and resistance are partly due to the viscosity of the fluid.

5. The suspension must be still or laminar, without turbulence in order not to alter the particle fall velocity, terminal velocity is attained as soon as settling begins.

2. Stokes' law and clay sedimentation

Several authors have reported that Stokes' law cannot adequately describe clay sedimentation in a column of fluid. In 1979, it was observed during experiments using organic mineral aggregates from lacustrine and marine environments that the settling characteristics of the aggregates in the range of 5 - 500μm were inconsistent with the assumptions of Stokes' law especially with smaller sizes of particles [1]. It has been shown that measured settling velocity of soil particles in dredged slurry is lower than that calculated using Stokes' law [2]. It was concluded that Stokes' law is not adequate to describe the settling velocity of clay particles with several reasons [3]. It is also noted that the settling velocity of reservoir and aquatic sediments do not comply with Stokes' law [4]. It was observed and concluded that the settling velocity of clays deviate from Stokes' law and as such suggested the introduction of a factor to bring the relationship into agreement with Stokes' law [5]. This factor is attributed to change in particle flocculated density, shape and size of flocculated particles. Several modifications of Stokes' law for settling velocity of sediments have been discussed [6]. It is therefore essential to find out why clay sedimentation deviates from obeying Stokes' law and this is examined by considering factors that affect clay sedimentation and relating them to assumptions under which the law was derived.

3. Factors in Clay sedimentation that defy Stokes' law

Several factors that affect clay sedimentation in fluids render Stokes' law inadequate for application in clay sedimentation because they inadvertently affect the settling velocity of clays in a fluid column. These factors include particle shape, size, concentration, flocculation, hindering velocity, water salinity, pH value, particle charges and presence of organic compounds [3, 7 - 8]. These issues have constituted limitations to application of the law in various situations including clay sedimentations because they were not considered. This has prompted a re-examination of the law and the assumptions associated with it in the light of factors that affect clay settling velocity in liquids.

3.1. Shape of Clay particles

Stokes' law assumes that particles are spherical in shape and smooth as shown in Fig. 1. However, clay particles are not smooth and spherical in shape; rather they are needle shaped, plate shaped or flaky. Clays are generally flat, small and with large surface area per weight. Kaolinite is made up of a series of layers of hexagonal shape like book pages as presented in Fig. 2. Montmorillonite is composed of small and thin particles as shown in Fig. 3, while the morphology of Illite particles is often flaky and thin on the edges as shown in Fig. 4. Kaolinite is migratory clay while Montmorillonite exhibits high shrinkage and swelling properties. The characteristics of Illite are classified between Kaolinite and Montmorillonite.

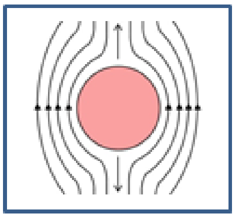

Fig. 1. Typical Particle Shape of Stokes' Law

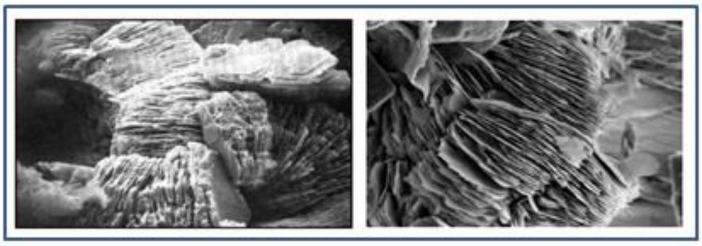

Fig. 2. SEM Shape Images of Kaolinite

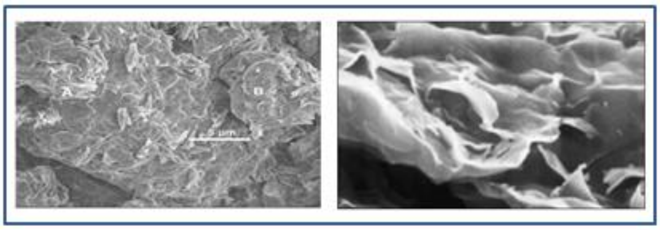

Fig. 3. SEM Shape Images of Montmorillonite

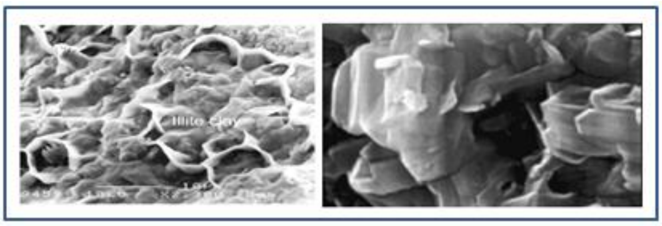

Fig. 4. SEM Shape Images of Illite

Studies have indicated that settling velocity decreases as the particle shape deviates away from sphericity [9], implying that Stokes' law will not give correct results for clay minerals. Clays therefore fall slower than spherical particles of the same mass due to the distorted spherical shape of their particles. Thus a modification factor is required to take care of shape and roundness factors. In some cases, new equations have been developed to capture the effect of shape factor on the settling velocity of particles in a fluid [10 – 12]. In fact in one equation, the shape factor for natural sediments is assumed to be 0.7 [13]. However in another case, the reduction in settling velocity caused by non-spherical shape of solid particles is quantified with an expression (Eq. 2) in velocity ratio [14] given as:

$$\xi = \frac{V_t}{V_{ts}}, \qquad \text{Eq. 2}$$

where

ξ = Shape factor of a solid particle (kg/m^3)

V_t = Terminal settling velocity of a non-spherical solid particle (m/s)

V_{ts} = Terminal settling velocity of a spherical solid particle (m/s)

3.2. Size of Clay particles

Fall velocity of particles also depends on particle size and density. The lower limit of particle size for validity of Stokes' law is 0.0002mm and the upper limit is 0.2mm. Brownian movement affects the settlement of particle sizes less than 0.0002mm. Some colloidal clay particle size dimensions are about 0.0002mm in size as shown in Table 1, hence exhibiting Brownian motion which distorts the settling velocity of such particles. Brownian movement of very small particles counteracts sedimentation. When particle density is lighter than the dispersion medium, it results in floatation and negative density difference. It has been pointed out that because of Brownian movement, Stokes' law cannot be applied to settlement of particles less than 0.1µm under gravity [15 – 16].

Table 1. Dimensions of Different Clay Particles

Clay Type	Length	Thickness
Kaolinite	0.3 – 3µm (0.0003 - 0.003mm)	0.005 – 1µm (0.000005 – 0.001mm)
Montmorillonite	0.1 – 1µm (0.0001- 0.001mm)	0.001 – 0.01µm (0.000001 – 0.00001mm)
Illite	0.1 – 2µm (0.0001 – 0.002mm)	0.01 – 0.2µm (0.00001 – 0.0002)

It has been stated that Stokes' settling does not apply to particles less than 5µm because small particles' electrostatic bonding, hydration and sorption of organic compounds reduce settling [17 - 18]. Experiments have also indicated that as particle size and volume increase, the settling velocity equally increases and vice versa. Equations have been developed to capture the effect of particle size in calculating the settling velocity of particles in a liquid and these equations express particle size in diameter [2, 11, 12, 19]. Additionally, different clay types have different particle densities; not the same density. It is assumed in the law that all the particles have approximately the same density. This means that the settling velocity of mixtures of clays cannot be determined using the law because different clay minerals have different densities and as such their fall velocity cannot be determined together.

3.3. Particle concentration and hindering velocity

The concentration of particles in a medium has a significant effect on settling velocity, hence falling clay particles are hindered and affected by other falling particles. It has been reported that at high sediment concentrations, particle settling velocities reduce to some small fraction of their clear water settling velocity. The hindered settling velocity of natural sand has been found to be higher than the hindered settling velocity of spheres of equivalent sizes. For fine and medium sands, the settling velocities are reduced to less than 20% of the clear water settling velocity [20]. In another research, it was observed that with increase in initial solid concentration, time of critical sedimentation reduced by approximately 50% [21]. It is also suggested that clay and silt content should be less than 5% by weight as sedimentation in more concentrated suspensions [16, 22]. The effect of concentration of particles in a liquid on the fall rate of individual particles has given rise to models of hindered settling velocity of particles [23].

Due to the fact that Stokes' law does not account for the effect of particle concentration on the fall rate of particles in a medium, efforts have been made to modify the equation to accommodate hindered settling velocity of falling particles. When a cloud of particles is settling in a medium under a laminar regime, the effect of other proximate particles hinders the settling velocity and this hindrance is dependent on the volumetric concentration of particles in the cloud [23]. The hindered settling velocity (Eq. 3) is hence expressed as:

$$V_{th} = V_t(1 - C_v)^m \qquad \text{Eq. 3}$$

where

V_{th} = Hindered settling velocity

V_t = Terminal velocity of the particle

C_v = Volumetric concentration

m = Empirical exponent related to the particle Reynolds number (Rep)

The value of the Richardson-Zaki index (m) is dependent on the range of value of the Reynolds number which is given on Table 2, where d stands for diameter. This equation was first modified by Ruby [24] who proposed an expression that considered all types of settling regime. Later, others also developed equations for the Reynolds number [25 – 26].

Table 2. Values of Richardson-Zaki Index with Reynold Number

$R_{ep} = V_t^d/v_f$	m
$R_{ep} \leq 0.2$	4.6
$0.2 < R_{ep} < 1$	$4.4\ R_{ep}^{-0.03}$
$1 < R_{ep} < 500$	$4.4\ R_{ep}^{-0.1}$
$500 \leq R_{ep}$	2.4

3.4. Particle interactions and flocculation

Often, there is interplay of forces when two colloidal particles approach each other in an entrained fluid during transportation. The theory of these forces is propounded by Derjaguin, Landau, Verwey and Overbeek termed the DLVO theory [27]. DLVO force is a sum total of the electric double layer, Van der Waals attraction and Born repulsion energies. Stokes' law assumes that there is no interaction between individual particles in the medium, which can be stated otherwise that a single settling particle has no influence from other particles in close proximity. However, DLVO attraction, repulsive forces of electric double layer forces and Van der Waals forces exist in clay particle colloid sedimentation and these forces cannot just be ignored. Equations that consider the effect of these forces and incorporate the equations of these forces into the settling velocity equations of clay minerals are necessary.

When clays are dispersed in a liquid medium, flocculation of particles is a common phenomenon. It occurs when dispersed individual particles in a medium collide with each other and stick together to form lager aggregates which is influenced by existing charges. Clay crystal surfaces carry negative charges, they form electric double layer and have the tendency to flocculate [28]. The fast falling particles therefore attracts and drags along with them other nearby finer particles especially those with opposite charges, thereby flocculating and forming large size aggregates. Flocculation is partly dependent on concentration of the particles in a medium. This is because increase in particle concentration increases settling velocity due to intensification of flocculation by an increase in the probability of particle collision [29 – 30]. The presence of organic compounds in a medium also affects the settling velocity of particles. Chase reported that the presence of dissolved organic matter in sea water and organic coatings on particles increased the settling velocity mostly for small particles, and a variation of about an order of magnitude was noted [1].

Particle flocculation phenomenon defies one of the assumptions associated with Stokes' law that the falling particles should not interact with other particles surrounding it. The settling velocity of reservoir and aquatic sediments that form aggregates therefore do not comply with Stokes' law and has necessitated modifications to suit particle flocculation. Models for settling of flocculated clay has been classified into six groups in the order of increasing complexity and these models have been presented and discussed [4].

3.5. Effect of water salinity and Ph value

Water salinity is another important factor that has effect on the settling velocity of fine particles. It has been noted that settling velocity increases with increase in salinity; in one set of experimental results, the settling velocities were observed to range from 0.3mm/s to 0.9mm/s ($3.0x10^{-4}$m/s to $9.0x10^{-4}$m/s). It was also stated that salinity increased cohesion of clay minerals, and cohesion increases flocculation tendency which increases the falling velocity of particles. In fact, it was reported that salinity has a direct relationship with the settling velocity of clay (Portela et al, 2013 and Sutherland et al). In another experiment where different concentrations of sodium chloride were used (0.5, 2, 4, 7 and 10ppt), it was observed that a decrease in sedimentation occurs from 0ppt to 4ppt, after which there was an increase in sedimentation as salinity increased up to a certain level (results indicate 7ppt) before sedimentation trend changed again either by remaining constant or increasing [21].

The settling velocity of kaolinite has been observed to decreases with increasing pH values from 3cm/min ($5.0x10^{-4}$m/s) at pH 3.5 to 1.4cm/min ($2.33x10^{-4}$m/s) at a pH value of 8.5 [31 – 32]. The pH values of various salts and at various concentrations for Montmorillonite, Illite and Kaolinite have been studied and generally, results show that pH value significantly affects settling velocity of clays in water [33]. The effects of salinity and pH value on clay sedimentation need to be modeled because they are not reflected in most equations that are used for determining the fall rate of clay particles in liquids. Even in Stokes' law these effects are neglected, rendering the law unfit for calculating the settling velocity of clays in natural settings such as in seawater, and in laboratories using saline water as a medium.

3.6. Other factors

Settling velocity regime is another important factor to consider when calculating the settling velocity of clays. Particle flocculation or aggregation result in larger particle sizes and particles sizes greater than 0.08mm diameter settle quicker and cause turbulence (a non-laminar regime). This necessitates that turbulent settling regimes be accounted for if Stokes' law must be used in determining the settling velocity of aggregated clay particles since the law was derived only for laminar regimes. Nevertheless, effort has been made to reflect flow regime in terms of Reynolds number [25 – 26].

According to experimental reports, temperature has considerable effect on the settling of particles in a fluid. In a certain study it was observed that with increase in temperature from 10°C to 45°C, critical sedimentation time decreased by about 49% [21]. It was speculated that increase in temperature increases stability of clay particles in suspension although the effect is much less evident than the effect of other physico-chemical factors [25]. It is also noted that flocculation of clay sediments increase with increasing temperature but there is a limit to this increase, which implies that settling velocity also increases with temperature but also has a limit [34]. Although Stokes' law assumes a constant temperature, it should be noted that in certain natural settings, maintaining a constant temperature in a long liquid column can pose a challenge.

4. Conclusion

Stokes' law is reliable and fundamental for modeling settling velocity of particles in a column of fluid under gravity within set limits. However, it has been pointed out that the settling velocity of clay particles under gravity in a fluid column does not obey Stokes' law due to various factors that affect clay sedimentation. These factors include particle shape, size, roundness, concentration, flocculation, hindrance velocity, salinity and pH value of the medium, particle charges and densities present in the medium. Stokes' law can therefore be modified to reflect these factors for clay settling velocity which constitute an area for further research, though efforts have been made to either modify the law or develop new models for particle settlement in fluids. Stokes' law of settling velocity for clays in a medium needs to be modified to capture factors that affect clay sedimentation which were not accounted for in the law. This area of research if successful will have a wider application and overcome the limitations of Stokes' law to clay settling velocity in a fluid column.

References

[1] Chase R.R.P., *Settling Behavior of Natural Aquatic Particulates*, Limnol. Oceanogr., **24**, 3, 1979, p.417 – 426.

[2] Zhang N., Zhu W., He H., Lv Y., Wang S., *Experimental Study on Settling Velocity of Soil Particles in Dredged Slurry*, Marine Georesources, Taylor and Francis Group, 35, **6**, 2017, p. 747 – 757.

[3] Berlamont J., Ockenden M. Toorman E., Winterwerp J., *The Characterization of Cohesive Sediment Properties*, Coastal Engineering, **21**, 1 – 3, 1993, p. 105 – 128.

[4] Floyd I.E., Smith S.J., Scott S.H., Brown G.L., *Flocculation and Settling Velocity Estimates for Reservoir Sedimentation Analysis*, US Army Corps og Engineers, ERDC/CHL CHETN-XIV-46, 2016, p. 1-10.

[5] Gibbs R.J., *Settling Velocity, Diameter and Density for Flocculants of Illite, Kaolinite and Montmorillonite*, Journal of Sedimentary Petrology, **55**, 1, 1985, p. 0065-0068.

[6] Shankar, M. S., Pandey, M., Shukla, A. K., *Analysis of Existing Equations for Calculating the Settling Velocity*, Water 2021, MDPI, **13**, 1987, p. 1 – 12.

[7] Wayne I. C., *Influence of Particle Density and Other Stokes' Law Assumption on the Precision and Accuracy of Particle Size Analysis*, Geological Society of America, **34**, 6, 2002, p. 207 – 307.

[8] Dietrich W. E., Settling Velocity of Natural Particles, Water Resources Reservoirs, **18**, 1982, p. 1615 – 1626.

[9] Komar P.D., Remers C.E, *Grain shape Effect on Settling Rates School of Oceanography*, Oregon State University, Corvallis, Journal of Geology, **86**, 1978, p. 193 – 209.

[10] Wu W., Wang S. S., *Formulas for Sediment Porosity and Settling Velocity*, Journal of Hydraulic Engineering, **133**, 2006, p. 229 – 233.

[11] Jimenez J.A., Madsen O.S.A., *A Simple Formula to Estimate Settling Velocity of Natural Sediments*, Journal of Water Port Coast, Ocean Engineering, **129**, 2003, p. 70 – 78.

[12] Camenen B., *Simple and General Formula for the Settling Velocity of Particles*, Journal of Hydraulic Engineering, **133**, 2007, p. 229 – 233.

[13] Cheng N. S., *Simplified Settling Velocity Formula for Sediment Particles*, Journal of Hydraulic Engineering, **123**, 1997, p. 149 – 152.

[14] Intermezzo I., *Settling Velocity of Solid Particles in a Liquid*, p. 1 – 12.

[15] Hongli Y., Minqiang F., Airong L., Lianping, D., *General Formulas for Drag Coefficient and Settling Velocity of Sphere based on Theoretical Law, Elsevier*, International Journal of Mining Science and Technology, **25**, 2015, p. 219 – 223.
[16] Stewart B.A., Christensen B.T., *Advance in soil science Boshland Texas*, USA, **20**, 1992, p. 24 – 25
[17] Salomone H. D., Olivieri N. A., Velz M. E., Raviola, L., *A Non-traditional Fluid Problem: Transition between Theoretical Models from Stokes' to Turbulent Flow*, European Physical Society, **39**, 2018, 035002, p. 1 – 16.
[18] Brun-Cottan J. C., *Stokes' Settling and Dissolution Rate Model for Marine Particles as a Function of Size Distribution*, Journal of Geophysical Research, **819**, 1976, p. 1601 – 1606.
[19] Rijn D.C.V., *Sediment of Transport, Part II: Suspended Load Transport*, Journal of Hydraulic Engineering, **110**, 1984, p. 1613 – 1641.
[20] Baldock T.E., Tomkins M.R., Nielsen P., Hughes M.G., *Settling Velocity of Sediments at High Concentrations*, Science Direct, Coastal Engineering, **51**, 2004, p. 91 – 100.
[21] Naghipour N., Ayyoubzadeh S. A., Sedighkia M., *Investigations on the Effect of Different Factors on Clay Particle Sedimentation in Fresh Water*', Journal of Biodiversity and Environmental Sciences (JBES), **5**, 2014, p. 75 – 81.
[22] Portela L. T; Ramos S. and Teixeira A.T., *Effect of Salinity on the Settling Velocity of Fine Sediments of a Harbor Basin*, Journal of Coastal Research, Portugal, **65**, 2013, p. 1188 - 1193.
[23] Richardson J.F., Zaki W.M., *Sedimentation and Fluidisation:* Part 1, trans. Instn Chem, Engrs, 1954, p. 32 – 35.
[24] Ruby W., *Settling Velocity of Gravel, Sand and Silt Particles*, American Journal of Science, **25**, 1933, p. 325 – 338.
[25] Soulsby R., *Dynamics of Marine Sands: A Manual for Practical Applications*, Thomas Telford: Telford, UK, 1997.
[26] Julien P. Y., *Erosion and Sedimentation*, Cambridge University Press, Cambridge, UK, 2010.
[27] Sokolov V. N., Tchistiakov, A. A., *Physico-Chemical Factors of Clay Particle Stability and Transport in Sandstone Porous Media*, European Geothermal Conference, Basel, Switzerland Proceedings, **2**, 1999, p.199-209.
[28] Venkatramaiah C., *Geotechnical Engineering, Revised Third Edition*, New Age International Publishes Ltd, India, Delhi, 1993, p. 50 – 51.
[29] Gombos M., Tall A., Trpcevska J., Kandra B., Pavelkova D., Balejcikova L., 2018), *Sedimentation Rate of Soil Microparticles*, Arabian Journal of Geoscience, **11**, 635, 2018, p. 1 - 10.
[30] Sutherland, B., Barett, K. J., Gingras, M. K., *Clay Settling in Fresh and Saline Water*, Environmental Fluid Mechanics.
[31] Chukwudi, Benneth C., *Settling Behavior of Kaolinite Clay in the Absence of Flocculants*, Department of Mechanical Engineering, Imo State University, Nigeria, The Pacific Journal of Science and Technology, **9**, 1, 2002, p. 212 – 218.
[32] Chaiwong N., Nuntiya A., Influence of pH, Electrolytes and Polymers on Flocculation of Kaolin Particles, Chiang Mai. J. Sci., **35**, 1, 2008, p. 11 – 16.
[33] Swineford Ada, *Clay and clay minerals*, Seventh National Conference on Clays and Minerals, State Geological Survey, University of Kansas, Pergamon Press, 1959, p. 17 – 61.
[34] Qiao G., Zhang J., Zhang Q., Feng X., Lu Y., Feng W., *The Influence of Temperature on the Bulk Settling of Cohesive Sediments in Still Water with the Lattice Boltzman Method*, Water MDPI, **11**, 945, 2019, p. 1 – 16.