Desalination and Water Treatment
www.deswater.com
doi no. 10.5004/dwt.2017.20423

72 (2017) 266–273
April

Coagulation and clarification of wastewater using rice husk ash

Chidozie Charles Nnaji*, Cordelia Nnennaya Mama, David Akachukwu Ezekoye, Izuchukwu Iwuchukwu

Department of Civil Engineering, University of Nigeria, Nsukka, Enugu State 410001, Nigeria, Tel. +234-8038948808, email: chidozie.nnaji@unn.edu.ng (C.C. Nnaji), Tel. +234-8063711674, email: cordelia.mama@unn.edu.ng (C.N. Mama), Tel. +234-9035834503, email: dakaezekoye@gmail.com (D.A. Ezekoye), Tel. +234-8066433319, email: izuchuksiwuh@yahoo.com (I. Iwuchukwu)

Received 4 August 2015; Accepted 6 December 2016

ABSTRACT

The coagulation ability of rice husk ash was investigated. Stormwater with suspended solids concentration of 540 mg/l was dosed with 500 mg/l, 800 mg/l, 1000 mg/l, 1200 mg/l and 1500 mg/l of rice husk ash at pH of 3, 5, 7 and 9. The samples were fitted into a jar test apparatus and stirred at 10 rpm for 30 min. Samples were drawn from the supernatant at intervals of 5, 10, 20 and 30 min and the suspended solids concentrations determined. Suspended solids removal was found to decrease with pH at rice husk ash concentration of 500 mg/l but increased with pH at rice husk concentrations of 800 mg/l to 1500 mg/l. Addition of 1500 mg/l to the raw water did not significantly improve the suspended solids removal efficiency over that obtained for rice husk ash concentration of 1200 mg/l. The maximum suspended solids removal was 93.34% for RHA concentration of 1500 mg/l at pH 9. An optimization process performed using kaolin yielded an optimum RHA concentration of 1500 mg/l at a pH of 3.0.

Keywords: Rice husk ash; Coagulation; Suspended solids; pH; Concentration

---

1. Introduction

Many coagulants are widely used in conventional water treatment processes for potable water production. Basically, coagulation is a process of addition coagulants to destabilize charged colloidal particles in water. Clarifying agents are used to remove suspended solids from liquids by inducing flocculation. Particles finer than 0.1 µm in water remains continuously in motion due to electrostatic charges (often negative) which causes them to repel each other. Once their electrostatic charge is neutralized by the use of chemicals, the finer particles start to collide and agglomerate under the influence of Van der Waal forces to form flocs. They can be classified as inorganic coagulant, synthetic organic polymer, and naturally occurring coagulants. Hydrolyzing metal salts, based on aluminum or iron, are very widely used as coagulants in water treatment. These salts bring about destabilization by adsorption and charge neutralization as well as by particle entrapment [1–4]. Coagulation process is influenced by a large number of factors which include water characteristics, chemical dose rates, pH, mixing conditions, flocculation times, the selection of chemicals and their order of addition, can all affect performance. Control of pH and alkalinity is also essential to maintain performance. Yan observed that pH is the most influential parameter affects non organic matter (NOM) removal from water [5].

Aluminum sulphate and polyaluminum chloride are the most widely used coagulants, because they are effective, readily available, and relatively inexpensive [6]. Ferric chloride has also been used extensively both at industrial and pilot scales. Alum was found to be effective coagulant in reducing solids, organics and nutrients in the dairy industry effluent for reuse in irrigation [7]. Konieczny et al. employed ferric chloride as a coagulant for food industry wastewater and recorded COD reductions of 77.3, 84.6

---

*Corresponding author.

and 48.8% for dairy, meat and fish processing wastewater respectively and BOD reductions of 85.4, 86.1 and 30.5% for dairy processing, meat processing and fish processing wastewater respectively [8]. Koohestanian et al. applied aluminum sulphate and ferric chloride as a coagulant and anionic polymer as coagulant aid [9]. Pernitsky and Edzwald present guidelines for the selection and use of polyaluminum chloride (PACl) coagulants and alum in terms of raw water quality and treatment method [10]. Annadurai et al. studied the simultaneous removal of turbidity and humic acid from high turbidity synthetic raw water using polyaluminum chloride (PACl) [11]. Synthetic polyelectrolytes are used as primary coagulant as well as coagulant aid to improve the strength of particle aggregates, enhance coagulation and deposition [12].

Despite the universal acceptance of aluminum sulphate as an effective coagulant, [13] noted that aluminum in coagulated drinking water has been regarded as a subject of human and environmental health concern. Driscoll et al. observed that the use of alum can increase the total Al (III) concentration from 0.37 ± 0.33 µmol/l in raw water to 1.8 ± 0.33 µmol/l in filtered water [14]. Dzulfakar et al. noted that chronic toxicity of aluminum in drinking water is associated with severe diseases of the nervous system such as Parkinson's dementia, amyotrophic lateral sclerosis and Alzheimer's disease [15]. Toxicity associated with accumulation of aluminum in water after coagulation has given rise to the search for nontoxic and inexpensive coagulant as replacement for alum and other synthetic coagulants. Natural coagulants have been successfully produced or extracted from microorganisms, animals or plants [16–18]. Ali et al. opined that *Moringa oleifera* is the best natural coagulant discovered yet, that can replace aluminum sulphate (alum) [19]. *Moringa oleifera* and *S. potatorum* seeds have been shown to concentrate the bio-active constituents which have coagulation activity for water clarification [20–22]. Santosa et al. reported that *Moringa oleifera* exhibits optimum coagulant activity in a protein concentration between 1 mg/L and 180 mg/L at pH 7.0 and that the two mechanisms involved are adsorption and neutralization of charges [23]. Al-Samawi and Shokrala successfully applied okra as a coagulant in wastewater treatment [24]. However, the coagulation ability of rice husk ash has not been extensively investigated. Rice husk ash has been successfully and efficiently used as an adsorbent for the removal of Fe^{+2} and Mn^{+2} [25], methylene blue and Congo red dye [26], Cr, Cd, Pb, Zn and Cu [27] and purification of bio-diesel [28]. It has also been employed as an efficient filter media [29] and as a partial replacement for cement in concrete production [30–32].

Rice husk accounts for 29% of the 649.7 million tons of rice produced annually worldwide [33]. The rice husk contains 80% organic volatile materials and remaining 20% silica. The chemical composition of the rice husk ash varies from rice husk to rice husk which may be due to geographical and climatic conditions, type of rice and the quantity of fertilizer used [34]. Its burning generates rice husk ash (RHA) which is rich in silica. Industrial importance of rice husk (RH) is due to the presence of silica in hydrated amorphous form. Ahmaruzzaman and Gupta [35] highlighted the presence of functional groups such as carboxyl, silanol and silicates in rice husk ash. The presence of these functional groups suggests that rice husk might possess coagulation properties. The objective of this work is to investigate the coagulation potentials of rice husk ash.

2. Methodology

2.1. Sample collection

The Rice Husk was obtained from a livestock kiosk at Beach Junction Nsukka, Enugu State Nigeria. The water sample was collected from a stormwater pond located along Achala Road, Nsukka. The water sample was brownish and comprises of solid particles, floating matter, colloidal particles. The floating matters were sieved out, leaving behind the water sample and its solid and colloidal particles.

2.2. Preparation of rice husk ash

The rice husk ash which was a whitish substance was obtained by ashing the rice husk for 5 h in 3 sets of 5 medium sized crucibles each in a furnace at a temperature of about 600°C and cooling in a desiccator for 2 h. The crucibles were weighed after cooling to determine the weight of the rice husk ash. The ash was sieved with sieve size 85 to obtain fine dusty particles of rice husk ash.

2.3. Settling column analysis

The raw water sample was stirred and then transferred into the settling column. The stop watch was immediately started and samples were collected from ports located at 20, 40, 60, 80, 100, 120 and 140 cm at intervals of 10, 20, 40, 60, 80 and 100 min. Total suspended solids are calculated from the weight of the particle residue on the filter paper gotten and measured after the filtration of the raw water sample and evaporation by oven drying of the water content of the wet filter paper. Suspended solids concentrations of samples were subsequently determined using standard methods (APHA method 2540D) [36].

2.4. Coagulation (Jar) test

The basic apparatus used for assessing coagulation performance is the jar test apparatus. One thousand millilitres (1000 ml) of raw water sample was measured into six 1000 ml beakers. The pH of the water samples were adjusted to 3. Five hundred (500), 800, 100, 1200 and 1500 mg/l of rice husk ash were weighed out using an electronic beam balance and then introduced into each of the beakers respectively. The paddles of the jar testing apparatus were inserted into the beakers and the jar test apparatus was turned at an angular velocity of 10 rpm was maintained for 30 min. At the end of the 30 min, the jar test apparatus was turned off; the beakers were brought out carefully to avoid disturbing the sample and allowed to settle. About 50 ml of the supernatants of the samples were taken from the four beakers at different time intervals of 5, 10, 20 and 30 min to determine suspended solids concentration. The procedure was repeated at different pH levels of 5, 7 and 9. To serve as control, the jar test was repeated for the raw water (without rice husk ash) at pH of 3, 5, 7 and 9. After the jar test, settled particles of raw water sediments and coagulated sediments were examined under a microscope.

2.5. Process optimization

In order to determine optimal conditions for coagulation of suspended solids by rice husk ash, an optimization scheme was designed using Minitab. A 2^3 randomized full factorial design with center point was employed. The parameters of the process investigated are pH, concentration of rice husk ash and stirring time. Kaolin was used for ease of control of suspended solids concentration. Stock solution was prepared by dispersing twenty grams (20 g) of kaolin in 1 L of distilled water, stirred for one hour and was allowed to stand for at least 24 h in order to achieve complete hydration. For each experimental run, 25 ml of stock kaolin suspension was introduced into 1 L of distilled water and the pH was then adjusted as required before introducing the rice husk ash. The mixture was stirred for the specified time duration and then allowed to stand for 30 min. The suspended solids concentration of the supernatant was thereafter determined. The response variable used was efficiency of suspended solids removal (R%).

3. Results and discussion

3.1. Settling column result and particles velocity distribution

Fig. 1 shows the result of the settling column analysis. The particles clearly exhibit the characteristics of colloidal particles. The suspended solids concentration of the raw water sample is 544 mg/m. After 10 min of settling under quiescent conditions, more than 50% of particles remained in suspension at a depth of 20 cm from the surface. However, about 96% of particles were removed after 100 min of settling. Plots and extrapolations of iso-removal lines have been presented in Fig. 1b. The plots suggest that the settling is Type 1 (discrete particles) settling rather than Type II (flocculent particles) settling. This is because for flocculent settling, the velocity of particles increases with depth as particles coalesce to form larger aggregates. However, for discrete particles, as the particles settle and the upper layer of the liquid is clarified, there is an accumulation of particles at the lower regions. This accumulation coupled with repulsion due to particle charges retard the velocity of the settling particle. This is clearly exhibited by Fig. 1b which shows that towards the bottom of the column, a longer time is required for particles to travel a unit distance under gravity. A further examination shows that each of the iso-removal lines approximately follows the relation:

$$H = \alpha Ln(t) + \beta \tag{1}$$

The values of α and β were determined for various removal efficiencies by regression and then plotted as shown in Fig. 2. Hence the specific expression for iso-removal lines was obtained as

Table 1
Experimental design for process optimization

Experimental run	pH	Concentration of RHA (g)	Stirring time (mins)
1	6	1	30
2	3	1.5	40
3	3	0.5	20
4	9	0.5	40
5	3	1.5	20
6	3	0.5	40
7	9	1.5	40
8	9	1.5	20
9	9	0.5	20

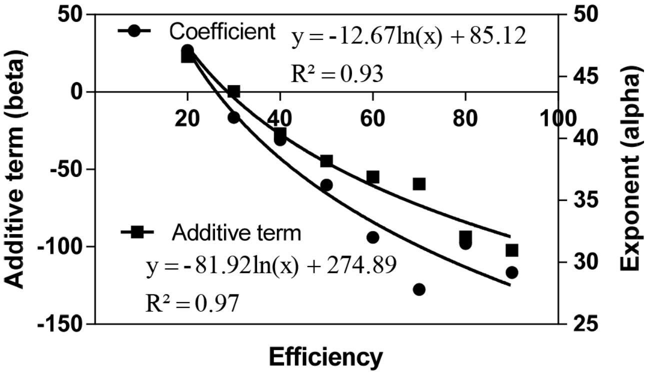

Fig. 2. Plots of Iso-removal lines constants.

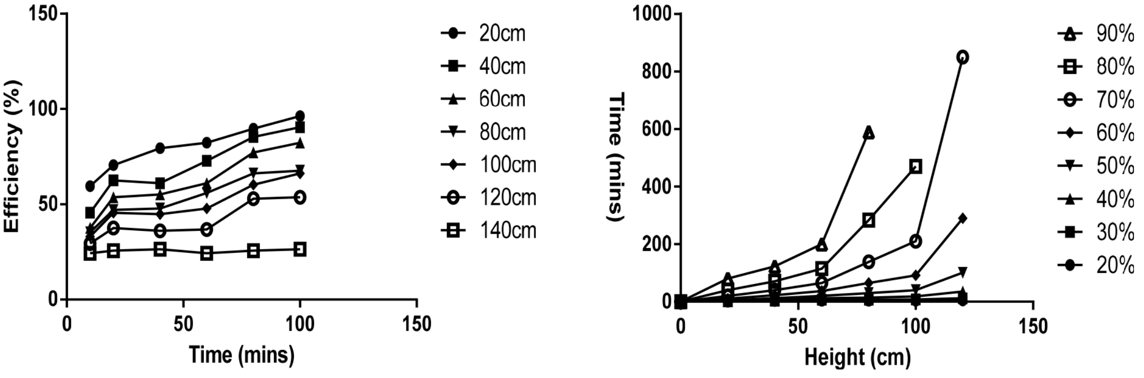

Fig. 1. (a) Unaided settling of raw water particles (b) Iso-removal plots of particles.

$$H = \left[85.12 - 12.67Ln(\eta)\right]Ln(t) - 81.92Ln(\eta) + 274.89 \quad (2)$$

where H is the depth, t is time and η is efficiency of suspended solids removal.

The particle settling velocity ranged from 0.12 m/h (0.002 m/min) to slightly above 8.4 m/h (0.14 m/min) as shown in Fig. 3. From Fig. 3, 80% of the particles have velocity less than 8.4 m/h.

3.2. Coagulation efficiency of rice husk ash

The addition of rice husk ash (RHA) to the raw water had significant effect on the particles settling. After five minutes of settling, the best efficiency of solids removal was as follows: 62.5% for 500 mg/l of RHA at pH 3; 69.12% for 800 mg/l of RHA at pH 7; 73.53% for 1000 mg/l of RHA at pH 3; 83.82% for 1200 mg/l at pH 7; and 84.56% for 1500 mg/l of RHA at pH 7. The maximum removal efficiency observed after 5 min of settling, without the addition of RHA was 43.53% at pH 9 (Fig. 4). This shows that the addition of RHA improved settling efficiency by about 40% within five minutes of settling. The optimum RHA concentration was 1200 mg/l as increase in RHA concentration to 1500 mg/l did not further improve settling efficiency irrespective of pH as clearly demonstrated by Fig. 4. In all cases, settling efficiency increased with settling time, however, about 84% of all suspended solids were removed within the first five minutes of settling.

The long term effect of RHA addition was demonstrated by plotting the removal efficiencies against RHA concentration after thirty minutes of settling (Fig. 5). Fig. 5 further shows that suspended solids removal efficiency increased linearly with RHA concentration from 500 mg/l to 1200 mg/l and thereafter no more increase in efficiency was observed. The highest increase in removal efficiency was observed from RHA concentration of 1000 mg/l to 1200 mg/l. This behavior can be explained as follows. When RHA is introduced into raw water, the ash quickly disperses into the water, engulfing the suspended particles and neutralizing the charges of the particles. RHA particles thus form a nucleus for the formation of larger RHA-suspended solids aggregates which are then quickly removed from suspension. Once adequate RHA has been added for the neutralization of particle charges, the excess RHA becomes redundant and will therefore not contribute to the coagulation process. The coagulation properties of RHA can be attributed to the presence of the hydroxyl, carboxyl and silanol functional groups and amorphous silica making it to act as a natural polyelectrolyte. From the Fourier transform infrared spectra (4000–400 cm^{-1}) conducted on the RHA sample, the following functional groups were identified: O–H stretching of carboxylic acids, alcohols and silanols with peaks at 2848.44 cm^{-1}, 3323.22 cm^{-1} and 3767.12 cm^{-1} respectively; the C–H stretching of alkanes and alkenes with peaks at 2842.44 cm^{-1} and 3014.42 cm^{-1} respectively; and the C=O stretching of carboxylic acids with peak at 1659.56 cm^{-1}. Adams et al. [37] confirmed the presence of alumino silicates and silica polymorphs due to O–Si–O and O–Al–O stretching modes as well as tetrahedral SiO_4 and crystalline christobalite in rice husk ash. Besides, [38] noted that RHA has a fibrous and longitudinal structure. This implies that RHA can achieve coagulation by two interwoven processes viz: particle charge modification and bridging flocculation. Bridging flocculation is a process where a substance adsorbs on separate particles and draw them together, resulting in the formation of flocs. Fig. 6 is a scanning electron micrograph of the both raw rice husk ash and flocs formed with suspended solids.

The performance of RHA as a coagulant was found to be dependent on the pH of raw water (Fig. 5b). At low concentrations (0–500 mg/l) efficiency of suspended solids removal generally increased with increase in pH. An increase in efficiency from 61.02 and 68.38% at pH 3 to 66.17 and 73.53% at pH 9 was observed for 0 and 500 mg/l of RHA respectively. However, at RHA concentrations above 500 mg/l, suspended solids removal efficiency decreased with increase in pH. Solids removal efficiency decreased from 79.41 to

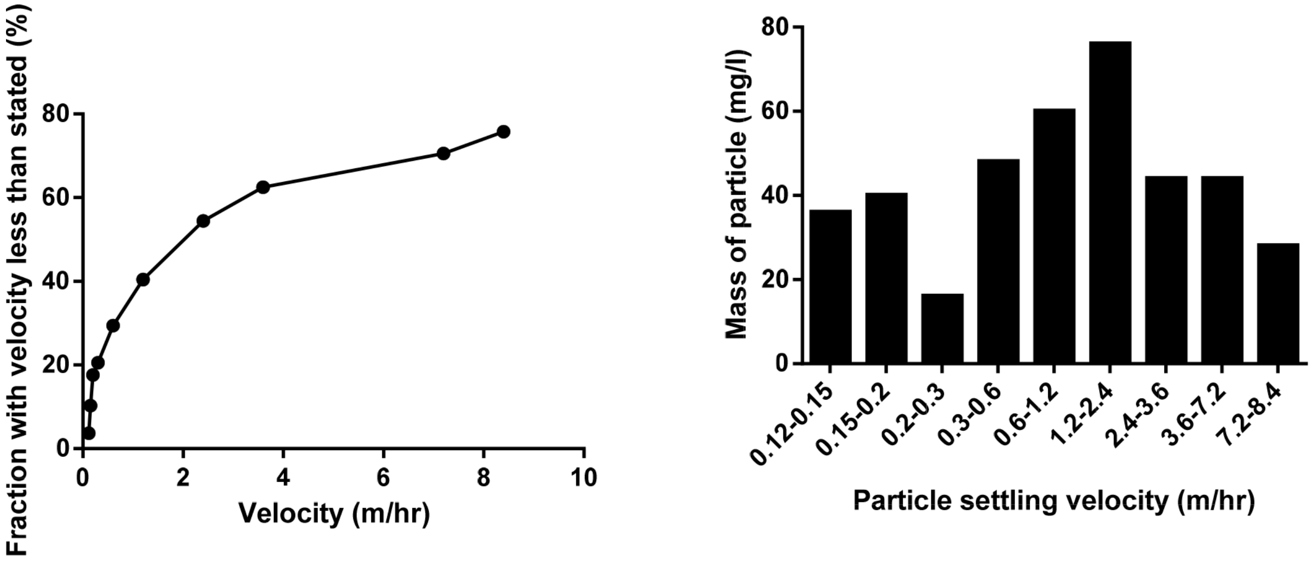

Fig. 3. (a) Velocity mass curve (b) Velocity distribution of particle.

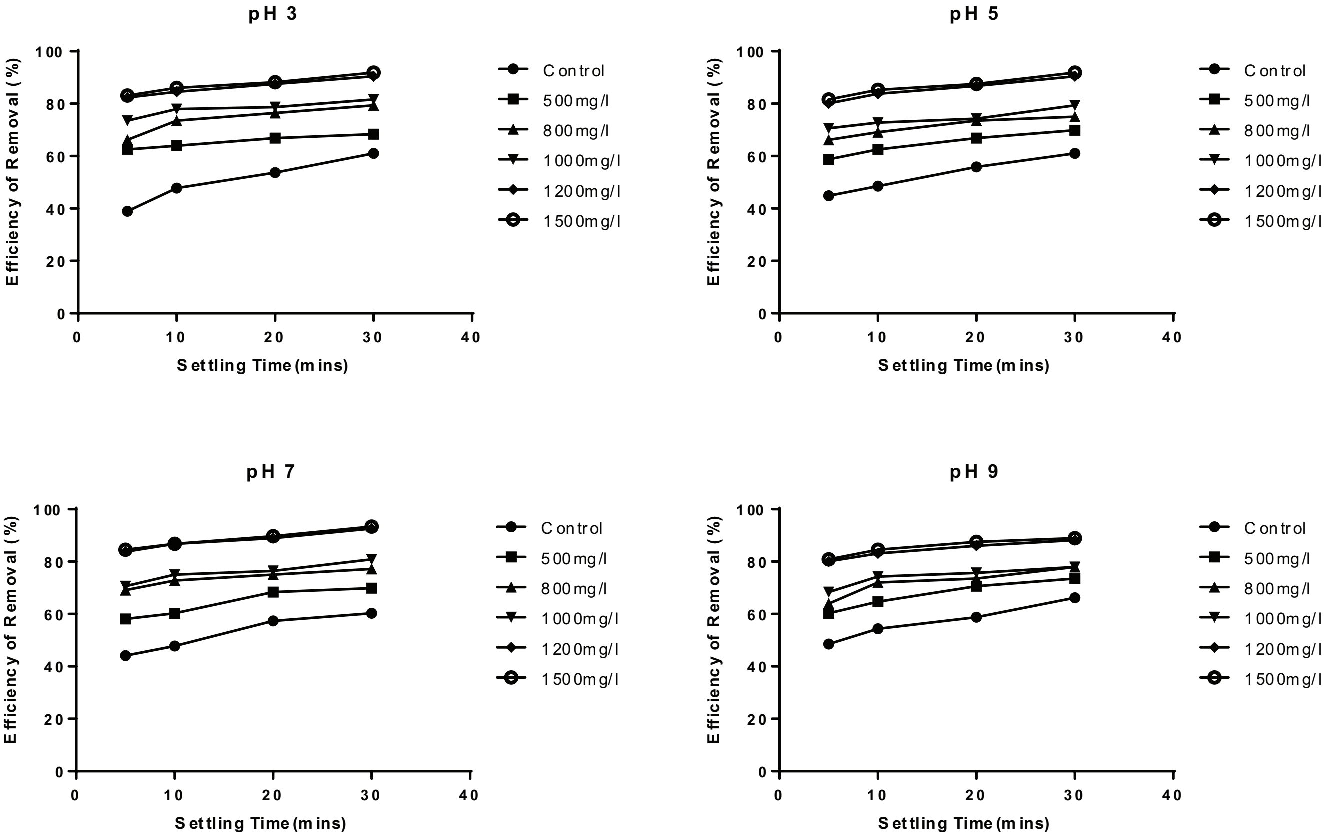

Fig. 4. Effect of rice husk ash concentration of suspended solids removal at different pH.

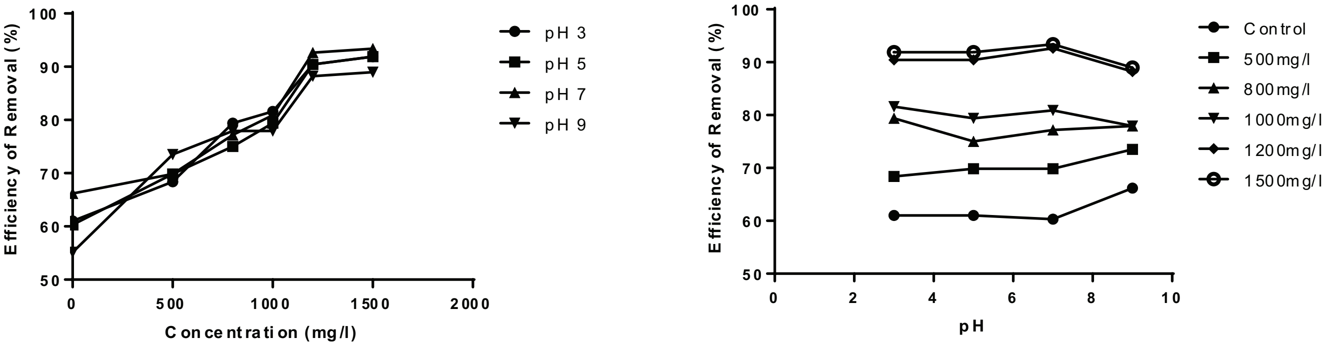

Fig. 5. Thirty minutes settling efficiency against (a) rice husk ash concentration and (b) pH.

77.41%, 81.62 to 77.94%, 90.44 to 88.24% and 91.91 to 88.97% RHA concentrations of 800, 1000, 1200 and 1500 mg/l respectively as pH increased from 3 to 9. The effect of pH on the efficiency of coagulation has been sufficiently established in the literature. Apostol et al. observed that optimum range of pH for coagulation using alum is from slightly acidic (6.5) to slightly basic (8.5) condition [39]. However, [40] established an optimum pH of 8. The results obtained from this study slightly agrees with the findings of [41] who found that optimum coagulation using alum was achieved in the acidic range between 3.5 and 5.5 and as pH increased, higher concentrations of coagulant were required for optimum coagulation. This simply means that efficiency of coagulation decreases as pH increases for high dosage of coagulant. Fig. 5b further shows that increase in pH to 9 has lower effect on suspended solids removal efficiencies of different concentrations of RHA. This is clearly portrayed by the progressive narrowing of in the spaces between the spaces in the plots of Fig. 5 from pH 3 to pH 9. It can be seen that at pH 9, removal efficiencies of 800 and 1000 mg/l of RHA merged at pH 9, while those of 1200 and 1500 mg/l merged at the same pH. This implies that lower (acidic) pH has more effect on suspended solids coagulation using RHA. It has been found that flocs are more stable at acidic pH, while basic pH facili-

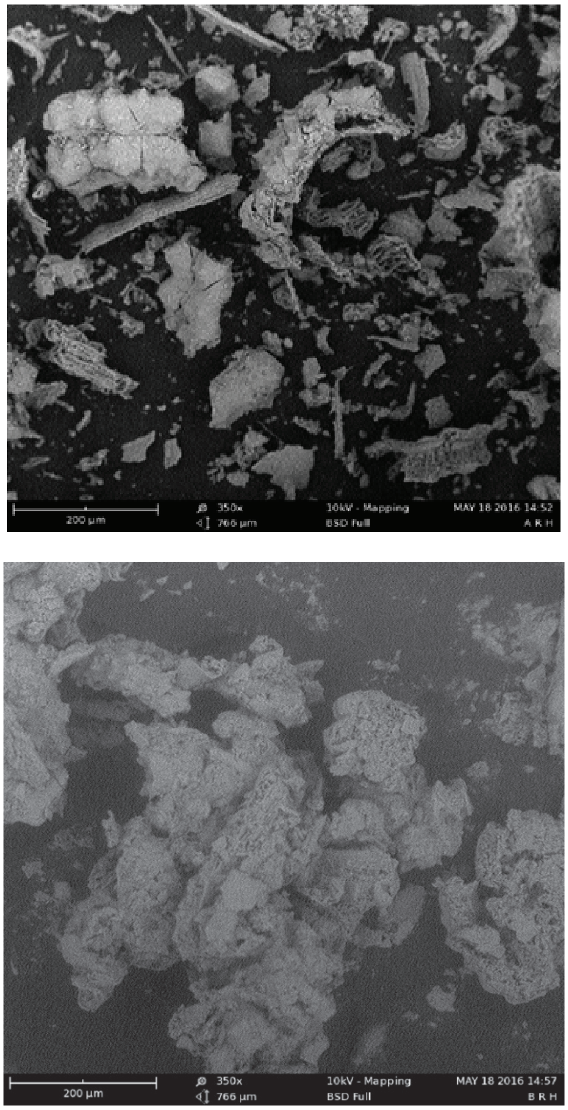

Fig. 6. SEM of (a) raw rice husk ash (b) coagulated particles with rice husk ash.

tates desorption of already entrapped particles thereby leading to a reduction in efficiency. Moreover, acidic pH results in the abundance of H^+ ions which impart a positive charge to the surface of RHA particles thereby enabling them to neutralize the negative charges of the suspended solids. Achadu et al. further observed that the interaction of RHA with anions is favoured at a pH less than that of the point zero charge [42]. They determined the point zero charge to occur at a pH of 8. The maximum suspended solids removal was 93.34% for RHA concentration of 1500 mg/l at pH 9. However, a removal efficiency of 92.65% was obtained for RHA concentration of 1200 mg/l at pH 7. Hence the excess 300 mg/l of RHA added above 1200 mg/l was redundant and can adversely affect suspended solids removal because the optimum condition had been exceeded. Yukselen and Gregory noted that excessive addition of coagulants can lead to charge reversal and subsequent destabilization of flocculated particles [43].

Table 2
Chemical composition of rice husk ash

Compound	Composition (%)
SiO_2	72.2
Al_2O_2	<0.001
P_2O_5	9.46
SO_3	<0.001
Na_2O	0.84
K_2O	1.7
CaO	2.94
MgO	0.8
TiO_2	0.645
Fe_2O_3	2.4

This study demonstrates that RHA can serve as effective coagulant in water and wastewater treatment. RHA is both a natural and nontoxic product from rice production and thus its use as a coagulant does not pose any health danger even if overdosed. Fadaei and Salehifar found that rice husk is an excellent source of protein, mineral and dietary fibre, hence allaying any possible fears of RHA toxicity [44]. By using natural coagulants, considerable savings in chemicals and sludge handling cost may be achieved. Al-Samawi and Shokrala reported that 50–90% of alum requirement could be saved when okra was used as a primary coagulant or coagulant aid [24]. Though the range of RHA concentrations required to achieve effective coagulation is orders of magnitude higher than that required for alum and ferric chloride, it is in the same range with that of *Moringa oleifera*seed. Othman et al. used *Moringa oleifera* concentrations ranging between 500 mg/l to 6000 mg/l for the coagulation of palm oil mill effluent [45]. Teh et al. found that up to 84.1% of TSS in agro-industrial wastewater can be removed by using 2000 mg/l of rice starch at pH of 3 [46]. Zin et al achieved only 12% suspended solids removal from partially stabilized landfill leachate using 2500 mg/l of Tapioca starch at a pH of 4 [47].

3.3. Optimization result

Fig. 7 shows the surface plots and interactions of the three parameters of interest. As earlier observed, the process is more favourable at low pH than at high pH. However, unlike the coagulation of particle in stormwater used in the first segment of the experiment, the coagulation of kaolin was found to be more sensitive to pH than concentration of rice husk ash (Fig. 7). Using the Minitab response optimizer function, the optimal conditions for coagulation using rice husk ash are pH 3.0, rice husk ash concentration of 1500 mg/l and stirring time of 40 min. Apart from the stirring time, these conditions correspond to those observed for the first phase of the experiment involving the use of stormwater. At these optimum conditions, the efficiency of suspended solids removal was 91.9% for stormwater as against 90.2% for kaolin suspension. It was however observed that the coagulation of suspended solids in stormwater using rice husk ash was more effective than the coagulation of kaolin using the same material. A careful examination of

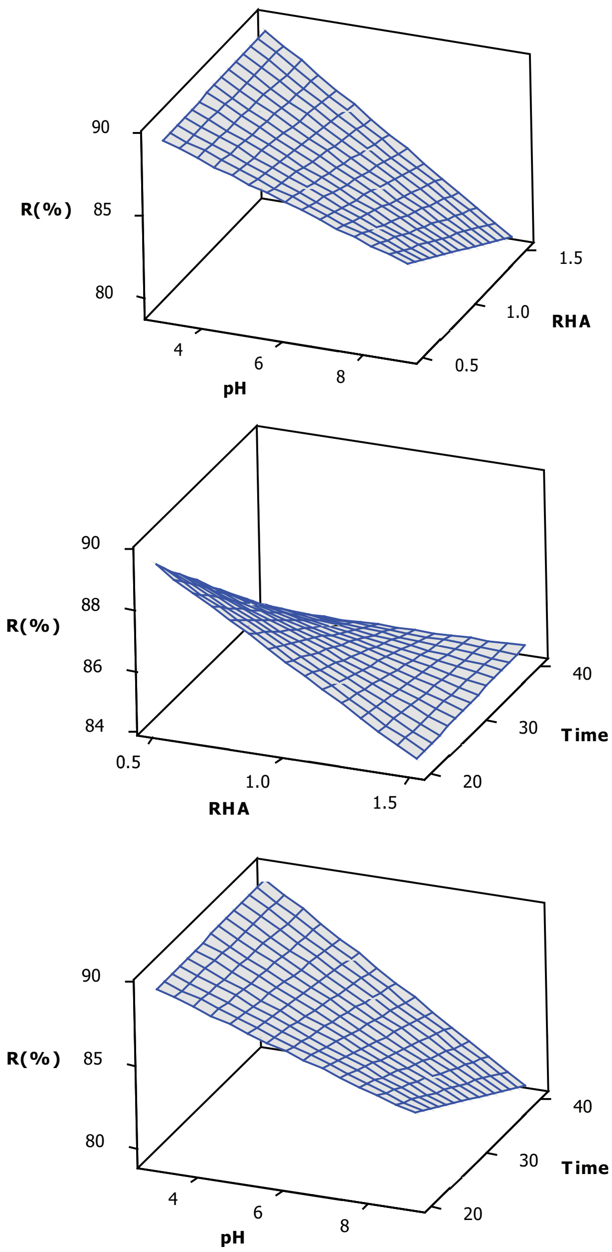

Fig. 7. Surface plots of variables.

the two processes also shows that suspended particles in stormwater are less sensitive to pH changes than kaolin. An analysis of variance performed on the data shows that addition of rice husk ash had significant effect on the kaolin suspension with an *F* value of 2305 for a critical *F* value of 4.49. The same applies to both stirring time and pH with *F* values of 218 and 1554 respectively. A linear regression model ($R^2 = 0.85$) describing the process was developed [Eq. (3)]. *R* is the efficiency of suspended solids removal, *C* is the concentration of rice husk ash (g) while *T* is the stirring time.

$$R(\%) = 79.96 + 3.25pH + 9.4C + 0.35T - 3.27pH * C - 0.122pH * T - 0.228C * T + 0.08pH * C * T \quad (3)$$

4. Conclusion

Rice husk ash was found to be an effective coagulant which can be successfully applied to water and wastewater treatment. Like every other coagulant used in water treatment, the coagulation efficiency of rice husk ash was found to be highly dependent on pH. At low rice husk ash concentration, coagulation efficiency increased with pH while at high concentration, coagulation efficiency increased as pH decreased. Generally, coagulation efficiency increased with increase in rice husk ash concentration. The optimum conditions for kaolin suspension coagulation by rice husk ash was found to be 1500 mg/l of rice husk ash, a stirring time of 40 min and a pH of 3.0.

References

[1] J. Duan, J. Gregory, Coagulation by hydrolyzing metal salts, Adv. Colloid Interface Sci., 100–102 (2003) 475–502.
[2] A. Amirtharajah, K. Mills, Rapid mix design for mechanisms of alum coagulation, J. AWWA, 74 (1982) 210–216.
[3] D. Bache, C. Johnson, E. Papavasilopoulos, F. McGilligan, Sweep coagulation: structures, mechanisms, and practice, J. Water Supply Res. T., 48 (1999) 201–210.
[4] B. Lartiges, J. Bottero, L. Derrendinger, B. Humbert, P. Tekely, H. Suty, Flocculation of colloidal silica with hydrolyzed aluminum: an 27Al solid state NMR investigation, Langmuir, 13 (1997) 147–152.
[5] M. Yan, D. Wang, J. Yu, J. Ni, M. Edwards, J. Qu, Enhanced coagulation with polyaluminum chlorides: Role of pH/alkalinity and speciation, Chemosphere, 71 (2008) 1665–1673.
[6] O. Sahu, P. Chaudhari, Review on chemical treatment of industrial wastewater, J. Appl. Sci. Environ. Manage, 17 (2013) 241–257.
[7] N. Al-Mutairi, M. Hamoda, I. Al-Ghusain, Coagulant selection and sludge conditioning in a slaughterhouse wastewater treatment plant, Biores. Technol., 95 (2004) 115–119.
[8] P. Konieczny, E. Ekner, W. Uchman, B. Kufel, Effective use of ferric sulfate in treatment of different food industry wastewater, Acta Sci. Pol., Technol. Aliment, 4 (2005) 123–132.
[9] A. Koohestanian, M. Hosseini, Z. Abbasaian, The separation method for removing of colloidal particles from raw water, Am. Eurasian J. Agric. Environ. Sci., 4 (2008) 266–273.
[10] D. Pernitsky, J. Edzwald, Selection of alum and polyaluminum coagulants: principles and applications, J. Water Supply Res. T., 55 (2006) 121–141.
[11] G. Annadurai, S. Sung, D. Lee, Simultaneous removal of turbidity and humic acid from high turbidity stormwater, Adv. Environ. Res., 8 (2004) 713–725.
[12] K. Vijay, M. Rubha, M. Manivasagan, B. Rames, P. Balaji, *Moringa Oleifera* - The nature's gift, Univers. J. Environ. Res. Technol, 2 (2012) 203–209.
[13] M. Guida, M. Mattei, C. Della Rocca, G. Melluso, S. Meric, Optimization of alum coagulation/flocculation for COD and TSS removal from five municipal wastewater, Desalination, 211 (2007) 113–127.
[14] C. Driscoll, D. Raymond, D. Letterman, Chemistry and fate of Al(III) in treated drinking water, J. Environ. Eng., 114 (1998) 21–37.
[15] M. Dzulfakar, S. Shaharuddin, A. Muhaimin, A. Syazwan, Risk assessment of aluminum in drinking water between two residential areas, Water, 3 (2011) 882–893.
[16] S. Muyibi, A. Alfugara, Treatment of surface water with *Moringa oleifera* seeds and alum a comparative study using a pilot scale water treatment plant, Int. J. Environ. Stud., 60 (2003) 617–626.
[17] H. Ganjidoust, K. Tatsumi, T. Yamagishi, R. Gholian, Effect of Synthetic and natural coagulant on lignin removal from pulp and paper wastewater, Water Sci. Technol., 35 (1997) 286–291.

[18] S. Kawamura, Effectiveness of natural polyelectrolytes in water treatment, J. AWWA, 83 (1991) 88–91.

[19] E. Ali, S. Muyibi, H. Salleh, A. Alam, M. Salleh, Natural coagulant from *Moringa oleifera* seed for application in treatment of low turbidity water, J. Water Resour. Prot., 2 (2010) 259–266.

[20] S. Muyibi, L. Evison, Optimizing physical parameters affecting coagulation of turbid water with *Moringa oleifera* seeds, Water Res., 29 (1995) 2689–2695.

[21] C. Ramamurthy, M. Maheswari, N. Selvaganabathy, M. Kumar, V. Sujatha, C. Thirunavukkarasu, Evaluation of eco-friendly coagulant from *Trigonella foenum-graecum* seed, Adv. Biol. Chem., 2 (2012) 58–63.

[22] R. Mohamed, A. Rahman, A. Kassim, *Moringaoleifera* and *Strychnos potatorum* seeds as natural coagulant compared with synthetic common coagulants in treating car wash wastewater: Case study 1, Asian J. Appl. Sci., 5 (2014) 693–700.

[23] A. Santosa, P. Paiva, J. Teixeira, A. Brito, L. Coelho, Coagulant properties of *Moringa oleifera* protein preparation: application to humic acid removal, Environ. Technol., 33 (2012) 69–75.

[24] A. Al-Samawi, E. Shokrala, An investigation into an indigenous natural coagulant. Environ. Sci. Health, A 31 (1996) 1881–1897.

[25] S. Abo-El-Enein, M. Eissa, A. Diafullah, M. Rizk, F. Mohamed, Removal of some heavy metals ions from wastewater by copolymer of iron and aluminum impregnated with active silica derived from rice husk ash, J. Hazard Mater., 172 (2009) 574–579.

[26] D. Sarkar, A. Bandyopadhyay, Adsorptive mass transport of dye on rice husk ash, J. Water Resour. Prot., 2 (2010) 424–431.

[27] M. Vieira, A. Almeida Neto, M. Silva, C. Carneiro, A. Melo Filho, Characterization and use of in natural and calcined rice husks for biosorption of heavy metals ions from aqueous effluents, Braz. J. Chem. Eng., 29 (2012) 619–633.

[28] M. Manique, C. Faccini, B. Onorevoli, E. Benvenutti, E. Caramão, Rice husk ash as an adsorbent for purifying biodiesel from waste frying oil, Fuel, 92 (2012) 56–61.

[29] J. Ijadunola, I. Adewumi, A. Ashaye, M. Oguntade, M. Ogunlade, Comparative study on the filtration properties of local sand, rice hull and rice hull ash, Sacha J. Environ. Stud., 1–2 (2011) 103–129.

[30] G. De Sensale, Strength development of concrete with rice-ash, Cement Concrete Comp., 28 (2006) 158–160.

[31] V. Saraswathy, H. Song, Corrosion performance of rice husk ash blended concrete. Const. Build. Mater., 21 (2007) 1779–1784.

[32] I. Obilade, Use of rice husk ash as partial replacement for cement in concrete, Int. J. Eng. Appl. Sci., 5 (2014) 11–16.

[33] G. Habeeb, H. Mahmud, Study on properties of rice husk ash and its use as cement replacement material, Mater. Res., 13 (2010) 185–190.

[34] V. Govindarao, Utilization of rice husk; A preliminary analysis, J. Sci. Ind. Res., 39 (1980) 495–515.

[35] M. Ahmaruzzaman, V. Gupta, Rice husk and its ash as low cost adsorbent in water and wastewater treatment, Ind. Eng. Chem. Res., 50 (2011) 13589–13613.

[36] APHA Standard methods for the examination of water and wastewater, 19th Edition. 2540 D, Total Suspended Solids Dried at 103–105°C, 1995, pp 2–57.

[37] F. Adams, B. Ikotun, D. Patrick, A. Mulaba-Bafubiandi, Characterization of rice hull ash and its performance in turbidity removal from water, Particul. Sci. Technol., 32 (2014) 329–333.

[38] M. Vieira, A. Almeida Neto, M. Silva, C. Carneiro, A. Melo Filho, Adsorption of lead and copper ions from aqueous effluents on rice husk ash in a dynamic system, Braz. J. Chem. Eng, 31 (2014) 519–529.

[39] G. Apostol, R. Kouachi, I. Constantinescu, Optimization of coagulation-flocculation process with aluminum sulfate based on response surface methodology, UPB. Sci. Bull., series b, 73 (2011) 1454–2331.

[40] S. Ramphal, M. Sibiya, Optimization of coagulation-flocculation parameters using a photometric dispersion analyser, Drink. Water Eng. Sci., 7 (2014) 73–82.

[41] E. Klimiuk, U. Filipkowska, A. Korzeniowska, Effects of pH and coagulant dosage on effectiveness of coagulation of reactive dyes from model wastewater by polyaluminium chloride (PAC), Pol. J. Environ. Stud., 8 (1999) 73–79.

[42] O. Achadu, O. Ayejuyo, F. Ako, C. Dalla, O. Olaoye, Adsorption of cadmium and lead ions from aqueous media by rice husk ash and sodium dodecyl sulfate combination, Int. J. Mod. Analyt. Sep. Sci., 3 (2014) 20–39.

[43] M. Yukselen, J. Gregory, The effect of rapid mixing on the break-up and re-formation of flocs, J. Chem. Technol. Biotechnol., 79 (2004) 782–788.

[44] V. Fadaei, M. Salehifar, Rice husk as a source of dietary fiber, Annals Biol. Res., 2 (2012) 1437–1442.

[45] Z. Othman, S. Bhatia, A. Ahmad, Influence of the settle ability parameters for palm oil mill effluent (POME) pretreatment by using *Moringa oleifera* seeds as an environmental friendly coagulant, J. Mater. Sci. Eng., 5 (2011) 332–340.

[46] C. Teh, T. Wu, J. Juan, Potential use of rice starch in coagulation-flocculation process of agro-industrial wastewater: Treatment performance and flocs characterization, Ecol. Eng., 71 (2014) 509–519.

[47] N. Zin, H. Aziz, S. Tajudin, Performance of Tapioca starch in removing suspended solids, colour and ammonia from real partially stabilized landfill leachate by coagulation-flocculation method, ARPN J. Eng. Appl. Sci., 11 (2016) 2543–2546.