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Kinetics and neuro-fuzzy soft computing modelling of river turbid water coag-flocculation using mango (*Mangifera indica*) kernel coagulant

Emmanuel Olusola Oke^a, Dauda Olurotimi Araromi^b, Lukuman Adekilekun Jimoda^b, and Jamiu Adetayo Adeniran^c

^aWater Treatment Process System Engineering Research Unit, Chemical Engineering Department, Michael Okpara University of Agriculture, Umudike, Nigeria; ^bChemical Engineering Department, Ladoko Akintola University of Technology, Ogbomosho, Nigeria; ^cChemical Engineering Department, University of Ilorin, Ilorin, Nigeria

ABSTRACT

This study investigates kinetics and Adaptive Neuro-Fuzzy Modeling (ANFM) of river turbid water coagulation-flocculation (CF) process using mango kernel coagulant (MKC). CF experiments were performed using jar test apparatus and the process kinetic-transport parameters (coagulation rate constant, half-life time, and particle diffusivity) were determined using kinetic-transport models. Grid-partitioning neuro-fuzzy programming codes were written and implemented in Matlab 9.2 software environment for the development of neuro-fuzzy architecture. The ANFM input data include initial water pH, initial water turbidity, biocoagulant dosage, CF time, and turbidity removal percentage (TRP) as output data. Generalized bell membership function was optimally selected for fuzzification of input variables and a hybrid algorithm was considered for the learning method of input-output data with constant output membership type. The minimum turbidity (0.51 NTU) of treated water was achieved at pH 12 and coagulant dosage of 2.5 mg/L with coagulation rate constant, half-life ($t_{1/2}$) and particle diffusivity 0.0194 s^{-1} , 10.01 min, and $7.267 \times 10^{-14} \text{ m}^2/\text{s}$, respectively. The correlation coefficient (R^2) between the experimental and neuro-fuzzy predicted values was 0.9924 and the ratio (K) of training error to testing error was 0.68. Thus, this study shows that ANFM can be used as a reliable tool for modeling river water CF and kinetic-transport parameter results are useful in process design, optimization, and control.

KEYWORDS

Kinetics; Mango kernel coagulant; Jar-test; Coagulation-flocculation; Neuro-fuzzy: Modeling

Introduction

Unavailability of clean water for drinking is a great challenge in most of developing countries of the world. Source of potable water such as surface river water is daily contaminated with different forms of particulate pollutants from industrial practices (Marichamy and Ramasamy, 2015; Amir et al., 2016). Consequently, river water contains suspended and dissolved particles with objectionable turbidity. Water treatment processes for particle removal can be achieved via sedimentation and filtration processes (Simate and Ndlovu, 2014). However, these unit operations cannot remove microscopically dispersed insoluble particles from water suspension due to particle interfacial tension that exists among the colloids in the suspension. The Coagulation-flocculation (CF)

process is one of the major pre-filtration processes used to remove microscopically suspended particles, objectionable color, turbidity, organic, and inorganic contaminants from industrial effluent (Huaili et al., 2011; Simate and Ndlovu, 2014; Marichamy and Ramasamy, 2015). In addition, the process is widely applied in water and wastewater treatment processes due to its inexpensive cost of installation and easy operation (Khouni et al., 2011; Menkiti et al., 2016).

A CF process is a physicochemical process which involves addition and dispersion of coagulant into the turbid water in order to destabilize the stable particles in suspension. The destabilization of turbid water is achieved by rapid mixing of the coagulant-turbid water system. As the coagulant is mixed with water at favorable conditions,

the micro-sized flocs become larger. Huaili et al. (2011); Khouni et al. (2011); and Menkiti et al. (2016) reported that coagulant dosage, initial water pH, initial water turbidity, flocculation settling time, and mixing speed influence efficiency of the process. The competency of the utilization of these coagulants is scientifically noteworthy. CF of turbid water has been earlier achieved using synthetic coagulants (Nnaji et al., 2014). Nevertheless, previous studies posited that the residual aluminum in treated water can provoke Alzheimer's disease in human health (Menkiti, 2007; Nnaji et al., 2014). In addition, it was well documented that synthetic coagulants produce high post-treatment sludge whose cost of sludge disposal is exorbitant (Menkiti and Onukwuli, 2012). Consequently, the use of natural coagulants has been proposed as safer substitute. Turbid water CF using natural coagulants is not detrimental to the human body and the sludge produced is environmentally biodegradable (Marichamy and Ramasamy, 2015). Therefore, the application of bio-coagulants has received great consideration in the scientific community. Previous works have shown that Moringa seed, bean seed, and other natural coagulants are efficient in the river and wastewater turbidity removal (Nnaji et al., 2014; Muthuraman and Sasikala, 2014; Marichamy and Ramasamy, 2015).

Mango (*Mangifera indica* L.) is a commercial tropical fruit in the world due to its nutritional content and quality. It is a fleshy stone fruit grown in almost all tropical and subtropical regions of the world. It ranks fifth in production among the major fruit crops and about 27 million tons is produced annually in the world (Olu-Owolabi et al., 2012). Nigeria ranks as the eight world's largest producer of mangoes producing 730,000 metric tons per annum (Olu-Owolabi et al., 2012). Mango seeds extensively contribute to the increase in solid waste generation and thereby increase the environmental burden. The properties and utilization of the mango kernel have been extensively studied in previous studies (Arogba, 1997, 1999a, 1999b, 2001, 2002a; Kittiphoom, 2012). The kernel has been utilized as a complementary ingredient in baking dough and biscuits (Arogba et al., 1998; Arogba, 2002b) and Arogba (2001, 2002a) also

reported that mango kernel contains polyphenol and high content of carbohydrate.

Previous studies reported that the conventional jar-test technique could not estimate the interactive effects of two process variables on turbid water biocoag-flocculation owing to non-linear relationship that exists among the process variables (Trinh and Kang, 2010; Chen et al., 2014). Response surface methodology (RSM) has been proposed as a suitable method for describing the non-linear behavior of turbid water CF (Trinh and Kang, 2010). In recent years, the applications of black box modelling (BBM) techniques have received great attention from scientists and engineers owing to the fact that the BBM requires only process data information for modelling complex engineering processes especially in chemical and petroleum process engineering (Zendehboudi et al., 2012; Ahmadi and Ebadi, 2014; Ahmadi and Bahadori, 2015; Oke et al., 2018). Artificial neural networks (ANN), adaptive neuro-fuzzy inference system or adaptive network-based fuzzy inference system (ANFIS), fuzzy inference system (FIS) and support vector machine (SVM) techniques in solving engineering process systems have also been used in previous studies for the prediction, estimation, and prognostics of process engineering systems (Ahmadi and Bahadori, 2015; Savic et al., 2016; Akbar et al., 2018). ANFIS technique is an integrated modeling method which combines the strength of ANN and FIS. ANFIS has been a potent tool for modeling and describing the non-linear relations among process variables. It is also widely used in process modeling, prediction, and forecasting (Melek and Avci, 2010; Amir et al., 2016).

Taleghani et al. (2017) reported that ANFIS is a simple tool for the estimation and prediction of the precipitated asphaltene, as a function of temperature, dilution ratio, and molecular weight of different n-alkanes, in a petroleum reservoir. The results obtained from the study were compared with computational correlation developed by Hu and Guo (2001). The findings show that ANFIS prediction is better than the existing models. Sahin and Gok (2016) also used ANFIS for the determination of wood drying rate; it was asserted that the estimation of ANFIS based technique is satisfactory, flexible, offering fast and simple solution

for modeling solid-liquid separation process. Generally, soft-computing modeling technique such as neuro-fuzzy is essential for studying process system, it allows process engineers to evaluate, analyze, simulate, and even forecast the behavior of a particular system (Amir et al., 2016).

Various studies have been conducted on bio-coag-flocculation of turbid water using a variety of bio-coagulants (Nnaji et al., 2014; Muthuraman and Sasikala, 2014; Okolo et al., 2016). However, information about the kinetic study of river turbid water CF using mango kernel coagulant (MKC) is scarce in the literature. Process kinetics study gives insight into the behavior of biocoag-flocculation at different operating conditions. The estimated turbid water biocoag-flocculation kinetic-transport parameters are useful in process design and optimization. In addition, there is a scarcity of research output on the application of exhaustive search and ANFIS techniques for modeling river turbid water CF. Exhaustive search technique is used for selecting the best set of process input variables that influence biocoag-flocculation process. Therefore, this study aims at bridging the existing gap in the literature by estimating kinetic-transport parameters and modeling of turbid water biocoag-flocculation using grid partitioning ANFIS technique.

Process kinetics and neuro-fuzzy model theory

CF kinetic theory

Equation 1 is the general CF kinetic rate equation of mono-dispersed particles (Nnaji et al., 2014; Menkiti et al., 2016).

$$\frac{-dN_t}{dt} = KN_t^n \quad (1)$$

N_t is the mass of particles per liter, t is the coagulation time, K is the n^{th} order process rate constant, and n is the order of the coagulation process. The negative sign shows that the total particles expressed in turbidity concentration decrease as a function of increasing time (t). The particle collision factor β_{BR} is for Brownian transport in turbid water CF system (Menkiti et al., 2016) and is expressed as Equation (2):

$$\beta_{BR} = 2K \quad (2)$$

The Smoluchowski's rate constant for rapid coagulation (K_R) is also given as Equation (3a):

$$K_R = \frac{4K_b T}{3\mu} \quad (3a)$$

The process rate constant (K) is a result of collision efficiency ε_p multiplied with the Smoluchowski rate constant for rapid coagulation (K_R). Therefore, collision factor for particles transport becomes Equation (3b).

$$K = \varepsilon_p K_R \quad (3b)$$

Equation 4 is derived from Equations (1–3):

$$\frac{-dN_t}{dt} = \frac{1}{2} \varepsilon_p K_R N_t^n \quad (4)$$

Brownian diffusion coefficient (D) of the particles due to concentration gradient is given as Equation (5):

$$D = \frac{K_b T}{B} \quad (5)$$

where $B = 6\pi\eta a$

T is the absolute temperature in Kelvin, D is particle diffusivity. For perikinetic aggregation of particles in coagulation/flocculation system, ' n ' theoretically in real practice is $1 \leq n \leq 2$ as reported (Menkiti, 2007).

For first order coagulation process, that is, $n = 1$; then, $\frac{-dN_t}{dt} = K_1 N_t$ upon an integration of this equation yields Equation (6):

$$\ln\left(\frac{N_0}{N_t}\right) = K_1 t \quad (6)$$

where N_0 is initial turbidity in mg/L, N_t is particle density/concentration at time t and K_1 is the first-order rate constant in (1/min). Equation (6) shows that the plot of $\ln\left(\frac{N_0}{N_t}\right)$ along y -axis and time (t) along x -axis should be a straight line passes through the origin and the line slope equivalent to K_1 . If coagulation process order is 2 i.e. ($n = 2$), rate expression in differential form for coagulation process is written as Equation (7):

$$\frac{dN_t}{dt} = N^2 k_2 t \quad (7)$$

where k_2 is second-order rate constant in l/(mg min). The second-order rate equation in integral form is written as Equation (8) after integration and simplification:

$$\frac{1}{N_t} = \frac{1}{N_0} + k_2 t \quad (8)$$

Equation (8) gives a straight line with slope k_2 and intercept made by the line along the y -axis is $\left(\frac{1}{N_i}\right)$ because the equation resembles slope-intercept form of straight line. One of the important timescales for the identification of the early stages in the coagulation process is the half-life period $t_{1/2}$. It is the time required for the initial turbidity to drop to one-half the original turbidity concentration. For first-order coagulation process, $t_{1/2}$ is shown by Equation (9):

$$t_{1/2} = \frac{\ln 2}{K_1} \quad (9)$$

For second order,

$$t_{1/2} = \frac{1}{(N_0 K_2)} \quad (10)$$

The particle distribution plot for coagulation/flocculation as a function of time can be generally represented as Equation (11):

$$\frac{N_{m(t)}}{N_0} = \frac{\left[\left(\frac{t}{2KN_0}\right)\right]^{m-1}}{\left[1 + \frac{t}{2(KN_0)}\right]^{m+1}} \quad (11)$$

Equation (11) can also be expressed as Equation (12):

$$\frac{N_{m(t)}}{N_0} = \frac{\left[\frac{t}{\tau}\right]^{m-1}}{\left[1 + \frac{t}{\tau}\right]^{m+1}} \quad (12)$$

Equation (12) gives generic solution of Smoluchowski's equation for particle size m -th order; thus,

For singlet particles ($m = 1$):

$$N_1 = N_0 \frac{1}{\left[1 + \frac{t}{\tau}\right]^2} \quad (13)$$

For doublet particles ($m = 2$):

$$N_2 = N_0 \frac{\left[\frac{t}{\tau}\right]^1}{\left[1 + \frac{t}{\tau}\right]^3} \quad (14)$$

For triplet particles ($m = 3$):

$$N_3 = N_0 \frac{\left[\frac{t}{\tau}\right]^2}{\left[1 + \frac{t}{\tau}\right]^4} \quad (15)$$

Equations (13–15) were used to determine particle cluster-size (singlet, doublet, and triplet size)

in bio-coagulation water treatment process using MKC's.

Exhaustive search and neuro-fuzzy model development

The 'exhsrchfunction' in Matlab environment was implemented in an exhaustive search within the available inputs of CF (initial water turbidity, initial water pH, settling time, and MKC dosage) to select the set of one, two, and three variable inputs combination that has the maximum influence on the turbidity removal percentage (TRP). Then, exhsrch function built a model for the combinations and trained. For neuro-fuzzy modeling, the pairs of the data were also trained and tested using grid partition ANFIS in fuzzy logic toolbox of Matlab 9.2 (2017a). The model for turbid water CF process was trained with the back-propagation method in combination with the least squares method, and various membership functions were tested during the training process. ANFIS network architecture consists of five layers as depicted in Figure 1. In Figure 1, square nodes (adaptive nodes) show that the parameters in these nodes are modifiable and to be learned, while the circle nodes (fixed nodes) shows that they are fixed parameters. A common rule set with two fuzzy if-then rules is as follows:

Rule 1: If x is A_1 and y is B_1 , then $f_1 = p_1x + q_1y + r_1$.

Rule 2: If x is A_2 and y is B_2 , then $f_2 = p_2x + q_2y + r_2$.

where A , B are linguistic terms that are user-defined representing a range of values. The sequence and functions of the layers are as follows:

Layer 1: Square node equipped with node function

$$O_i^L = \mu_{A_i}(x) \quad (16)$$

Assuming x and y be the two typical input values fed at the two input nodes, which then transforms those values to the input membership functions. Where O_i^L is the membership function of A_i and x is the input parameter to the node.

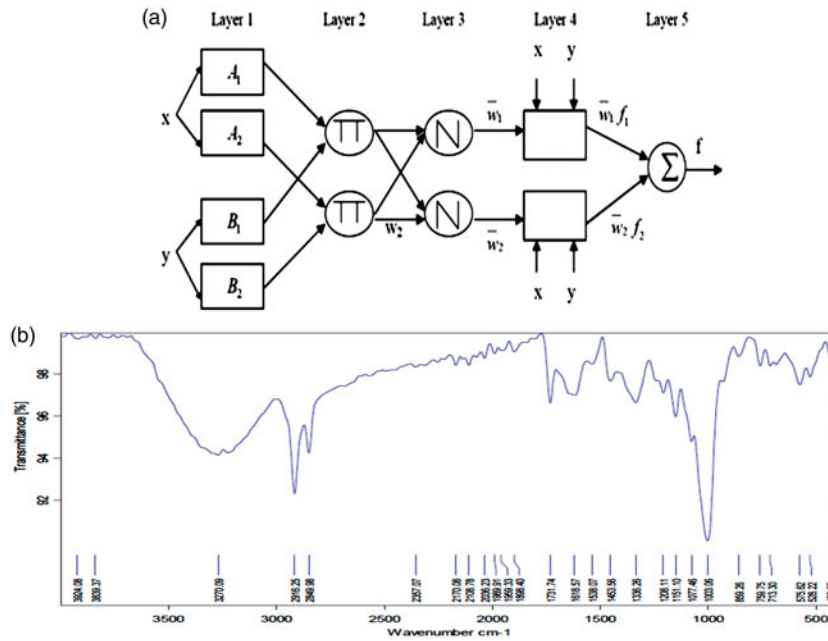


Figure 1. Equivalent ANFIS structure (Source: Chiu, 1994) (a). FTIR spectra of mango kernel powder (b).

A_i is the linguistic label connected with the node function.

Layer 2: This node increases the homeward bound signal and releases the product out of the layer.

$$w_i = \mu_{A_i}(x) \times \mu_{B_i}(y) \text{ and } i = 1, 2 \quad (17)$$

Layer 3: Circle node - node calculates the ratio of i-th rule's firing strength to the sum of all rules firing strengths:

$$w'_i = \frac{w_i}{w_1 + w_2}, \quad i = 1, 2 \quad (18)$$

Layer 4: Square node with node function:

$$O_i^4 = w'_i f_i = w'_i (p_i x + q_i y + r_i) \quad (19)$$

where p , q , and r are parameter set (consequent, linear parameters).

Layer 5: Circle node—This node computes the overall output as summation of all incoming signals.

$$O_i^5 = \text{overall output} = \sum_i w'_i f_i = \frac{\sum_i w_i f_i}{\sum_i w_i} \quad (20)$$

Neuro-fuzzy modelling performance index

The Neuro-fuzzy network performance was appraised and evaluated by using statistical criteria and K value (K value relates the ratio of training and testing error of the network). The value

of K close to one shows the degree of predictability and dependability of ANFIS structure (Huaili et al., 2011). Equations (21) and (22) show the equations used to calculate correlation coefficient (R^2) and K value, respectively.

$$R^2 = \frac{n(\sum xy) - (\sum x)(\sum y)}{\sqrt{[n \sum x^2 - (\sum x)^2][n \sum y^2 - (\sum y)^2]}} \quad (21)$$

$$K = \frac{\text{Checking error}}{\text{Training error}} \quad (22)$$

Artificial neural network modelling

Artificial neural network (ANN) architecture was developed in Matlab 9.2 (2017a) software environment (MathWorks, Inc., Natick, MA). A three layer feed-forward back propagation neural network consists of input, output, and intermediate hidden layers with tangent sigmoid transfer function at the hidden layer and a linear transfer function at the output layer were used for modeling of the TRP of turbid water biocoag-flocculation. The input layer corresponds to the four experimental parameters including the initial water turbidity (NTU), initial water pH, settling time (minute), MKC dosage (mg/s), and contact time (min). The output layer was the TPR (%). All data derived from the CF experiments were

divided into training, validation, and test sets with a ratio of 70, 15, and 15%, respectively.

Material collection, preparation, and characterization

River turbid water

The sample of river water dam used was obtained from Ogbomoso waterworks treatment plant, located in Ogbomoso, Oyo State, Nigeria was collected. The characteristics of the turbid water sample in are presented in Table 1.

Mango kernel coagulant preparation

Mango seed biomass was collected from Mango tree plantation at Baptist Theological Seminary, Ogbomoso, Oyo State, Nigeria. Mango kernels were removed from the seed, dried, crushed into fine powder and then sieved through a 30 μ m mesh. The powder was deoiled by solid-liquid extraction using soxhlet extraction chamber (Electro Thermal Soxhlet apparatus, England – Model-13789). The processed mango kernel powder cake residue was obtained and proximate composition of the sample is given in Table 2. Percentage suspension of mango kernel powder was prepared and thoroughly shaken for 30 min using the magnetic stirrer to enhance water extraction of the coagulating agent in order to obtain MKC stock solution (Okolo et al., 2016).

Crude mango powder characterization

Functional groups in the processed powder were obtained by Fourier transform infrared (FTIR) technique using an FTIR Spectrophotometer (model Tensor27 Germany). Surface morphology of the MKC powder was also examined using scanning electron microscope (SEM; model VPFESEM Surpra 35VP).

Batch CF experiment

The coagulation/flocculation experiments were carried out using pilot program-controlled jar test apparatus (Pilot-Scale flocculator, Model CL-4) at room temperature. Dosages of MKC extract in the range 0.5–2.5 mg/L were added directly to

Table 1. Characteristics of Ogbomoso waterworks river water.

Parameter	Value
Turbidity	3.7–32.5 NTU
pH	6.2–8.9
Total dissolved solid	56–74 mg/L 4 mg/L
Electrical conductivity	114–159 $\frac{\mu}{m}$
Temperature	27–29 °C

Table 2. Proximate composition characterization of MKC.

Parameter	Value
Moisture (%)	14.5
Protein (%)	7.1
Carbohydrate (%)	67
Fat (%)	5.9
Ash (%)	4.2
Crude fiber (%)	1.3

500 ml of turbid water, at room temperature (27.5 °C). The suspension adjusted to pH range of 3–12 using HCl and NaOH, were subjected to 2 min of rapid mixing (250 rpm), 18 min of slow mixing followed by 30 min of settling. Then, the clarified river turbid water was withdrawn using pipette from 2 cm depth from the beaker to measure residual turbidity. Turbidity removal percentage (TRP) is expressed as Equation (23):

$$\text{TRP} = \frac{N_o - N_t}{N_o} \times 100 \quad (23)$$

Results and discussion

Mango kernel powder characterization

Figure 1 shows the FTIR spectrum of Mango Kernel Powder (MKP) and it revealed that the sample contained complex molecules. It was observed that many sharp-strong bands are present. The sharp-strong band observed within the characteristic peak around 3473 cm^{-1} corresponds to the hydroxyl group, and the 3300–3200 cm^{-1} band corresponds to the amine group in the analysis of the spectrum represented in Figure 1(a). The characteristic peaks obtained from this investigation (MKP FTIR characterization) compared with the results of previous study are similar (Rai et al., 2016). Hydroxyl and amine groups present in biomass possess coagulating/flocculating property (Zhang et al., 2010). As reflected in spectra, MKC contains the necessary characteristics that have enhanced its coagulating/flocculating potential. Previous studies have reported this

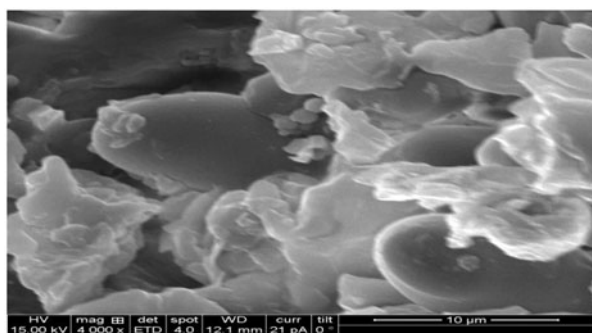


Figure 2. SEM micrograph of MKC (magnification 4000×)

assertion (Li et al., 2008; Zhang et al., 2010). However, the strong hydrogen bond and O-H group found in MKC could also be responsible for better coagulating/flocculating potential for the treatment of turbid water. Other functional groups such as C-H and C-O may not contribute much to coagulation-flocculation process based on previous findings (Prasertsan et al., 2006; Hanafiah et al., 2007; Li et al., 2008; Zhang et al., 2010). Figure 2 shows surface morphology characteristics of MKC using SEM at 4000 magnification power. A surface characteristic found in MKC is similar to the previous investigation (Li et al., 2011; Jhumur et al., 2016). Figure 2 indicated the presence of circular polymeric starch granules in the sample.

Effect of MKC dosage on TRP

Figure 3(a) shows the effect of MKC dosage on TRP at different initial water pH during river turbid water CF. Figure 3(a) revealed the highest turbidity removal of 96.8% during the process, while the least removal was 34%. The highest and lowest turbidity removals correspond to 2.5 and 0.5 mg/L, respectively as shown in the Figure 3(a). It was also observed that as the dosage increases, the rate TRP increases. The trend observed from this investigation is similar to a previous study (Nnaji et al., 2014). The turbidity removal efficiency obtained from this study agrees with the results obtained in similar study on water deturbidization (Menkiti et al., 2017). This result confirms that increase in MKC dosage in river turbid water system increases CF property of turbid water system and thereby increases the rate of destabilization of the particles in

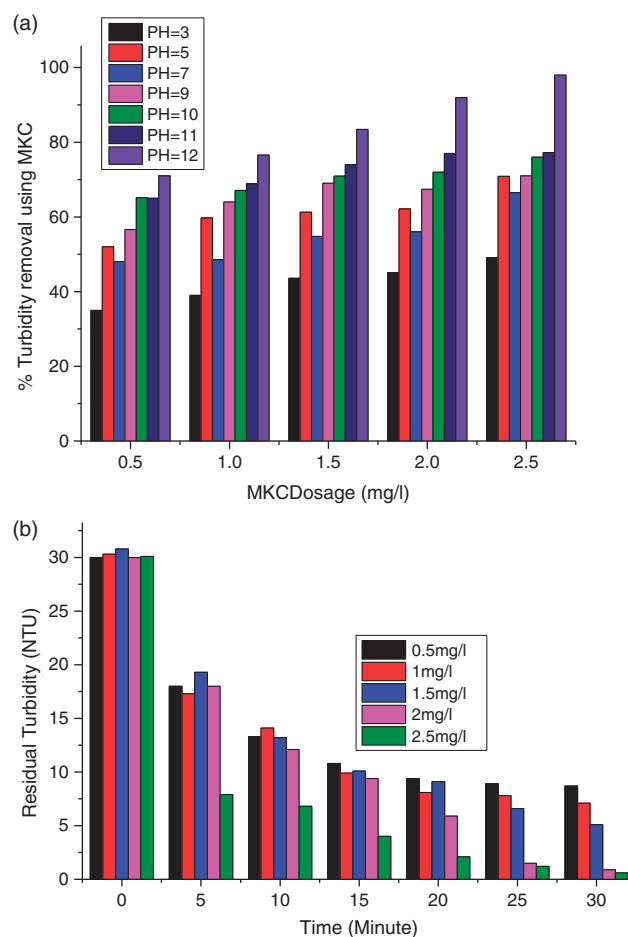


Figure 3. (a) Coag-flocculation efficiency using varying coagulant dosage and initial pH. (b) Coag-flocculation kinetics at varying coagulant dosage.

suspension. Moreover, Figure 3(a) shows that 2.5 mg/L of MKC for turbid water treatment has highest turbidity removal for all the pH values. This result confirms that at 2.5 mg/L dosage, the buoyant force against particle settling during the treatment might have been swept away. Figure 3(b) shows the change of residual turbidity as a function of CF process time for various dosages of MKC. The linear relationship between turbid water turbidity concentration and process time for all MKC doses is presented in Figure 3(b). The process time progresses for all the doses and the residual turbidity concentration decreases. This occurrence confirms that river turbid water CF, using MKC as bio-coagulant, is a time-dependent process. This observation is in tandem with the observation of previous biocoag-flocculation studies (Marichamy and Ramasamy, 2015; Menkiti et al., 2016).

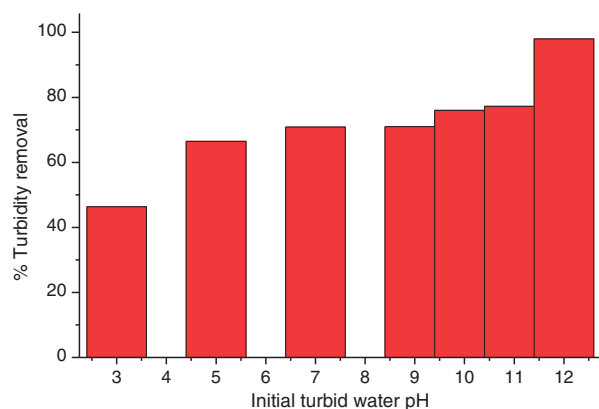


Figure 4. Variation of initial turbid water pH on the efficiency at coagulant dosage 2.5 mg/L.

Effect of initial water pH on TRP

The influence of river turbid water initial pH on CF efficiency at the predetermined optimum dosage is presented in Figure 4. The turbid water was adjusted in the range of 3–12 through 0.5 mol/L HCl and NaOH solutions. The initial water pH influenced CF of turbid water as shown in Figure 4. With increase in initial turbid water pH in the acidic environment, the removal was not appreciable. However, turbidity removal in the alkaline environment clearly increased as the alkalinity of initial water increased. This shows the high dependence of turbid water turbidity removal on the initial water pH and MKC can destabilize more stable particles in the alkaline environment than the acidic. Little and thin flocs were formed in acidic pH values while better and visible flocs were formed at alkaline initial pH water. The best turbidity removal was achieved at initial water pH 12 and 2.5 mg/L dosage. The dependence of turbidity removal efficiency observed in this work at alkaline medium has been observed in a previous study (Nnaji et al., 2014).

River turbid water CF kinetics

To investigate CF kinetics of river turbid water, the residual turbidity of river turbid water was monitored at different process periods of time at predetermined optimum initial water pH and coagulant dosage (Figure 5). The second order CF rate equation was used to test the experimental data and to explain the process kinetics. The

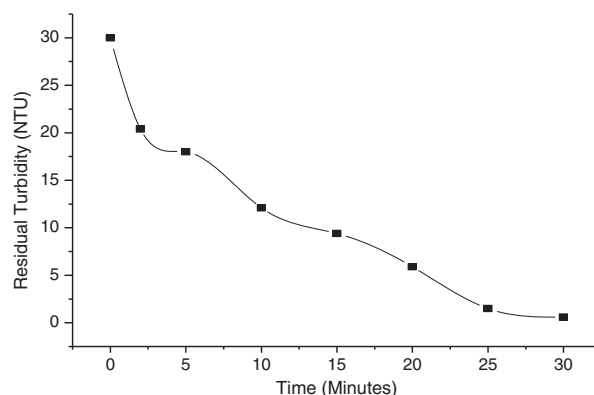


Figure 5. Kinetics of residual turbidity at initial water pH 12 and 2.5 mg/L of coagulant dosage.

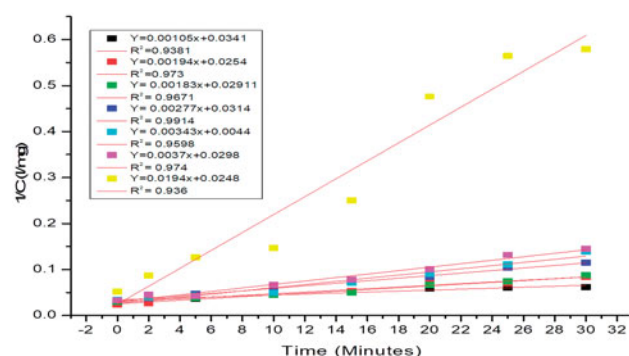


Figure 6. Kinetic plot of turbid water coag-flocculation at coagulant dosage of 2.5 mg/L at different pH values.

integral form of rate equation is represented in Equation (8). Figure 6 shows the plot of turbidity concentration against various pH values at 2.5 mg/L. It was observed that the obtained data agreed with the second-order kinetic equation. The coefficient of determination (R^2) was used to determine the extent of linearity of the plot. Figure 6 show the degree of linearity and the linearity of plot is confirmed by R^2 values.

The second order kinetic and transport parameters of turbid water CF are presented and evaluated in Table 3. The CF rate constant (K), the coefficient of determination (R^2), R^2 -adjusted ($\text{adj } R^2$), Pearson value and sum of square error (SSE) were obtained from fitting of experimental data to Equation (8) (Table 3). As pH of river water increased the rate constant increased. Also, the minimum and maximum rates constant at 2.5 mg/L MKC were 0.00105 for pH 3 and 0.0194 for pH 12, respectively. This shows that the rate of CF of river turbid water was highest at pH 12.

Table 3. Turbid water Coag-flocculation kinetic-transport parameters for varying pH with Constant Dosage of 2.5 mg/L.

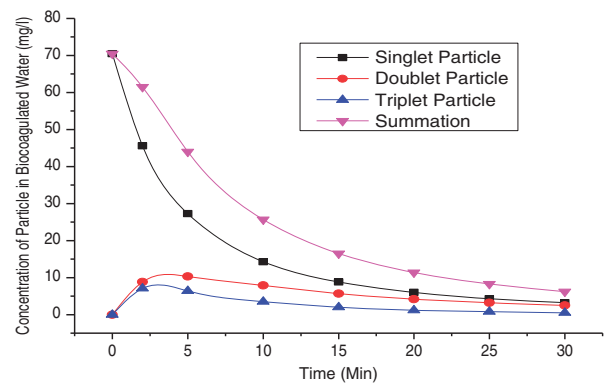
Parameter	pH = 3	pH = 5	pH = 7	pH = 9	pH = 10	pH = 11	pH = 12
R^2	0.9381	0.973	0.9671	0.9914	0.9598	0.9746	0.9364
Adj R^2	0.9251	0.981	0.9583	0.98631	0.94264	0.96907	0.92259
Pearson r	0.93815	0.942	0.96718	0.99141	0.95985	0.97464	0.98277
SSE	1.02E-04	1.10E-04	1.27E-04	9.72E-05	2.64E-04	2.29E-04	5.28E-04
K	0.00105	0.00194	0.00183	0.00277	0.00343	0.00377	0.0194
B_R	0.0021	0.00288	0.00366	0.00554	0.00686	0.00754	0.0289
D	5.16E-14	5.21E-14	5.54E-14	5.96E-14	5.96E-14	5.27E-13	7.26E-13
Half life	29.9855	15.5279	12.245	11.5741	11.44	10.7181	10.01
K_R	1.2E-17	1.21E-17	1.21E-17	1.17E-17	1.17E-17	1.17E-17	1.14E-17

The process half-life time suggests the time in which the dissolved and colloidal particles reduce to half of the initial water turbidity. The observed trend of half-life time of the process decrease as the rate constant increase, suggests that inverse proportional relationship exists between the two kinetic parameters. The trend and pattern observed from this study conforms to a previous study (Marichamy and Ramasamy, 2015).

Particle diffusivity or particle diffusion coefficient (D) is a transport property that determines the rate of particle movement or transfer during the treatment. It was observed from Table 3 that as rate constant increases, particle diffusion coefficient increases. This implies that as the rate of destabilization of stable particle increases, it invariably increases the rate of transport of destabilized particles during the process. The behavior of particle diffusion coefficient in this study is consistent with a previous study (Jimoda et al., 2013). The adequacy and reliability of the fitting were evaluated with statistical parameters such as R^2 , adj R^2 , SSE, and Pearson value. The value of R^2 , adj R^2 , and Pearson for validating the fitting ranges from 0.922 to 0.992. The obtained values are very high and close to unity. This indicates that the fit is in good agreement with the turbid water CF data.

Particle size distribution estimation

Equations (13–15) were used to predict the rate of particle aggregation during the treatment at different levels, that is, singlet, doublet, and triplet. The time evolution of formation of doublet and triplet particles is presented in Figure 7. The singlet and total particles decrease more quickly than the doublet and triplet as indicated in Figure 7. This occurs as a result of the rapid transformation of colloidal particles into micro-

**Figure 7.** Particle size distribution in turbid water-mango kernel coagulant system of 2.5 mg/L for pH = 12.

sized flocs during the process. Consequently, as flocculation continues, the doublet and triplet particles settle down. It was observed that doublet and triplet particles reached the peak at 4.6 min and finally, the triplet, doublet, and singlet particles attained equilibrium at 28 min. It was also observed that all the particles reached equilibrium almost at the same time. The pattern observed in this result is similar to a previous study (Menkiti et al., 2016).

ANFIS simulation result

Effect of input variables on TPR as output

The exhaustive search function in Matlab used root mean square error (RMSE) as the parameter for measuring the degree of influence of each or combined variables on the output of the process (Olajide et al., 2014). Figures (8–10) show the effect of one and combined (two or three combined variables) input variables on the output (TRP) of the turbid water CF. Figure 8 shows the effect of each variable on TRP of CF. The second left-most input variable (CD: coagulant dosage) in Figure 8 has the least RMSE which shows the

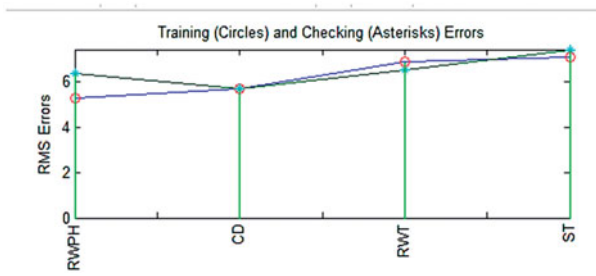


Figure 8. One input variable's influence on turbidity removal percentage.

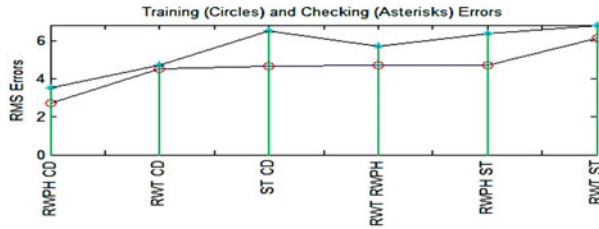


Figure 9. All two input variable combinations and their influence on turbidity removal percentage.

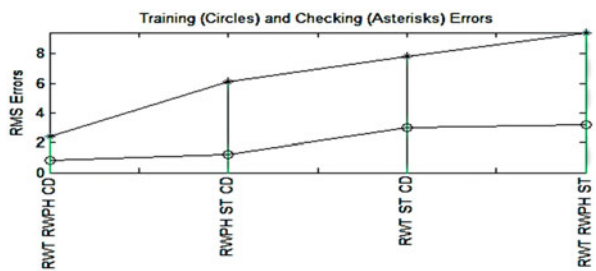


Figure 10. All three input variable combinations and their influence on turbidity removal percentage.

most significant contribution to the output (TPR). The plot (Figure 8) from the function (exshrc) clearly indicates that the input variable 'coagulant dosage' with RMSE of 5.6978 is the most influential factor affecting the TPR. However, the order of the significance of other variables is indicated according to the value of the training error (RMSE). Thus, the order of the variables is as follows: influence of raw water pH is greater than raw water turbidity i.e. raw water pH > raw water turbidity > settling time. The plotting of training and checking errors is analogous, which suggests that there is no overfitting during exhaustive search process. This suggests that further higher variable combination can be investigated.

To verify the optimal combination of two inputs that result in the minimal training error, exshrc function was also used to search for the optimal combination of two input variables. The results, as indicated in Figure 9, show that 'raw water pH (RWPH)' and 'coagulant dosage (CD)' form the optimal combination of two input variables with 3.5183 RMSE. The order of the variable combination is as follows: (raw water pH and coagulant dosage) > (raw water turbidity and coagulant dosage) > (raw water turbidity and raw water pH) > (raw water pH and settling time) > (settling time and coagulant dosage) > (raw water turbidity and settling time). For further investigation, three variable combinations were explored for building the exhaustive model. Figure 10 demonstrates the result of selecting three variables inputs, in which 'raw water pH', 'coagulant dosage', and 'raw water turbidity' as the best combination of three input variables. However, the minimal training (and checking) errors do not reduce from that of the best two-input model, which indicates that the newly added variable 'raw water turbidity' improves the prediction. The sensitivity analysis results of input to output variables in this study are analogous to existing investigation (Mashaly and Alazba, *in press*). Cosine amplitude method was used in the earlier study but the results look alike. This confirms the exhaustive search method as a potential tool for input-output variables sensitivity technique.

Input variables setting for ANFIS architecture

ANFIS model prediction depends on the number of certain parameters which include the number of input membership function (IMF), type of input and output membership functions and epoch number (Amir et al., 2016). To obtain the best prediction of turbid water biocoag-flocculation, the developed ANFIS structure was simulated at various conditions such as IMF of gauss mf, gauss2 mf, gbell mf, tri mf, trap mf, psig mf, and dsig mf as well as epoch numbers (EN) from 10 to 500. The correlation coefficient (R^2) and the ratio (K) of training error and testing error were used to evaluate the degree of reliability of the network. The preliminary simulation study

Table 4. ANFIS model Simulation for 'constant' output membership function.

NIMF	Type of IMF	Epochs	R^2	K	Computational time
2	gbell	500	0.9926	0.68	4 s
2	tri	500	0.970	0.34	4 s
2	gauss	500	0.981	0.42	4.1s
2	gauss2	500	0.982	0.33	4 s
2	pi	500	0.986	0.34	5.2 s
2	dsig	500	0.986	0.41	4 s
2	psig	500	0.9861	0.51	4 s

NIMF: number of input membership function; IMF: input membership function.

was conducted in order to obtain optimum epoch number needed for ANFIS setting 500 epoch value was considered as the best epochs for the subsequent simulation.

Table 4 shows the simulation result of constant output membership function type at different input membership function type. R^2 , K , and simulation computational time were the parameters used for the goodness of the neuro-fuzzy network. It was observed from Table 4 that the highest and the lowest computation times were 5.2 and 4.0 s, respectively. The results of ANFIS simulation revealed that the values of R^2 and K ranged from 0.981–0.9926 and 0.33–0.68, respectively. The R^2 values for different ANFIS architectures in Table 4 are comparable with previous studies (Amir et al., 2016; Mashaly and Alazba, in press; Oke et al., 2018). Therefore, the results are consistent with existing findings. It is from the same table that gbell input membership function has the highest values of R^2 (0.9926) and K (0.68) with relatively low computational time.

Similar simulation results were presented in Table 5; however, 'linear' output membership function type was tested on the ANFIS network. It was noticed from Table 5 that the highest and the lowest computation times were 4.1 and 4.0 s, respectively. The values of R^2 and K ranged from 0.997–0.839 and 0.591–0.340. It was observed from Table 5 that tri input membership function has the highest R^2 value of 0.997 and K (0.578) with relatively low computational time. However, the training and checking curves of the data are characterized by high degree of over-fitting.

The simulation results of Tables 4 and 5 were compared in order to select the best input membership type for further investigation. Based on the high values of K and R^2 observed in tri and gbell membership functions in Tables 4 and 5,

Table 5. ANFIS model simulation for linear output membership function type.

IMF	Type of IMF	Epochs	R^2	K	Computational time
2	gbell	500	0.902	0.591	4 s
2	tri	500	0.997	0.578	4 s
2	gauss	500	0.839	0.35	4.1s
2	gauss2	500	0.619	0.34	4s
2	pi	500	0.933	0.36	4.2s
2	dsig	500	0.935	0.45	4s
2	psig	500	0.935	0.48	4s

NIMF: number of input membership function; IMF: input membership function.

Table 6. ANFIS model Simulation for gbell and tri input membership function.

IMF	Type of time IMF	TOMF	R^2	K	Computational
3	gbell	linear	0.718	0.52	4.2 min
3	gbell	constant	0.937	0.57	4.2 min
3	tri	linear	0.902	0.38	5 min
3	tri	constant	0.619	0.39	4 min

TOMF: type of output membership function.

Table 7. Best ANFIS model setting for river turbid water bio-coag-flocculation.

Parameters	Types/values
Input membership function	gbell
Output membership function	Constant
Epochs	500
Optimization method	Hybrid
Number of input MF	2

the two MF were used for simulating the network with three (3) input membership functions as shown in Table 6. The computational times in Table 6 are high as compared with Tables 4 and 5. It was observed from Table 6 that gbell and constant input-output membership functions type gave highest R^2 and K values with reasonable computational time. Based on the foregoing, Table 7 shows the optimum parameters (two input gbell MFs, constant output MF type, and 500 epochs) for the biocoag-flocculation neuro-fuzzy network.

Figure 11 shows ANFIS model structure for river turbid water CF using the setting parameters in Table 7. The structure consists of 16 rules and one output as indicated in Figure 11. It was observed that the network is simple because it also consists of 16 linear and 24 non-linear parameters as shown in Figure 11. Figure 12 compares the predicted results with the experimental data; it was observed that correlation

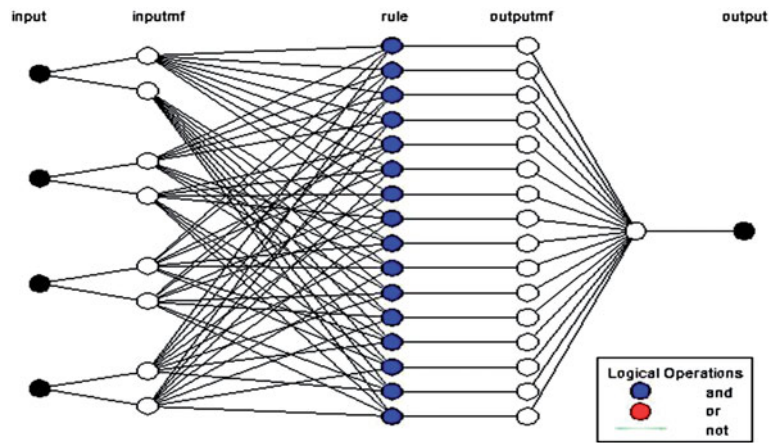


Figure 11. Neuro-fuzzy network with two gbell MF inputs and constant output MF type.

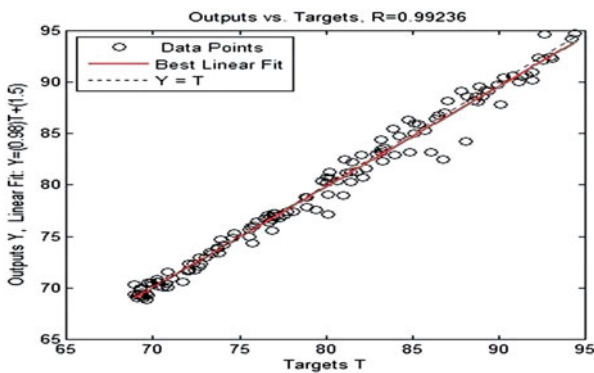


Figure 12. The graphical output of ANFIS outputs plotted versus the corresponding targets.

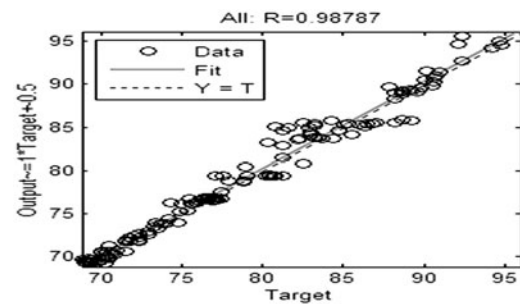


Figure 14. The graphical output of the network outputs plotted versus the corresponding targets for the Levenberg-Marquardt algorithm.

ANFIS result compared with ANN result

The optimal structure of the ANN model was determined based on the minimum MSE value obtained for the network training and its prediction. The training was stopped after 21 iterations for the Levenberg-Marquardt algorithm – (LMA) because the differences between training error and validation error started to increase. Figure 13 illustrates training; validation and test mean squared errors for the LMA. A regression analysis of the network response between ANN outputs and the corresponding targets was performed. The graphical output of the network outputs plotted versus the targets as shown in Figure 14. Considering the non-linear dependence of the data, linear regression shows a good agreement between ANN outputs (predicted data) and the corresponding targets (experimental data). The best linear fit was indicated by a solid line and R^2 is 0.9878. The ratio (K value) of training error to testing error, as shown in Figure 13, is

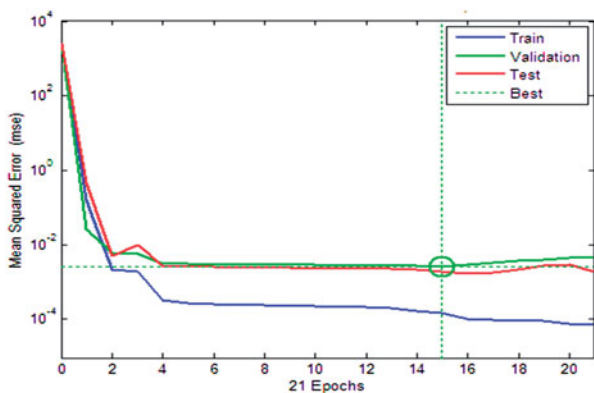


Figure 13. Training, validation, and test mean squared errors for the Levenberg-Marquardt algorithm.

coefficient (0.99236) is very high as evident in Figure 11. The high value of R^2 indicates that the network predicts river turbid water CF accurately. Hence, the predicted results obtained from this study showed that grid partitioning ANFIS model can simulate turbid water Cf process using MKC.

0.7. ANFIS and ANN techniques were compared in this study and it was noticed that both soft-computing techniques predicted turbid water bio-coag-flocculation satisfactory. It was observed that the regression coefficients (R^2) for both methods are very close to one, indicating the correlation between experimental and soft-computing predicted data is high. Both the ratio (K value) of training error to testing error is also close to each other. However, ANFIS R^2 is higher than the value obtained for ANN network. Based on the comparison, ANFIS model was found superior to the ANN.

Conclusion

CF of river turbid water with MKC shows that minimum turbidity value of treated water (from 30 to 0.51 NTU) was achieved at pH 12 and coagulant dosage of 2.5mg/L with coagulation rate constant, $t_{1/2}$, and particle diffusivity 0.0194 s^{-1} , 10.01 min, and $7.267 \times 10^{-14}\text{ m}^2/\text{s}$, respectively. This investigation has established that CF of river turbid water using MKC is influenced by initial water pH, coagulant dosage, and coagulation/flocculation time. Thus, the process is coagulant dosage, CF time, and pH-dependent process. This study has also indicated that the coagulant dosage is the most influential factor affecting the process and combination of coagulant dosage and initial water pH has effect on turbidity removal percentage. Combination of raw water pH, coagulant dosage, and raw water turbidity are also the best combination of three input variables for the best removal. R^2 between the experimental and neuro-fuzzy predicted values were 0.99236 and the ratio (K) of training error and testing error was 0.68; therefore, ANFM can be used as a reliable tool for modeling river water CF.

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