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MACHINE LEARNING MODEL TO DETERMINE THE CURVE-FITTING EQUATION FOR PREDICTION OF THE START-UP OF NON-BROWNIAN SUSPENSIONS IN NON-NEWTONIAN FLUID

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Abstract. *The rheological behavior of complex fluids such as non-Brownian suspensions is currently a required topic in the cosmetic, food, and petroleum industries. Since in processes where flow start-ups occur, high pumping pressures are necessary to exceed the gel strength. The interactions between the particles are one of the main problems for correctly obtaining the rheological properties of the non-Brownian suspension, especially in viscoelastic matrices with yield stress. Therefore, it is necessary to understand the transition from solid to liquid phase that occurs during flow start-ups in non-Brownian suspensions since variables such as the fraction of solids and particle size can influence the measurements. In the current work, controlled shear rate rheometric tests were conducted to investigate the yielding of Carbopol® gel solution with different spherical particles of sizes between 20 and 50 µm. The creep tests are performed with different constant shear stress values. A model based on machine learning is proposed, where the shear rate is considered. In order to find the best-supervised fit of the data obtained through rheological measurements. The analysis and adjustment of curves through machine learning allows to have a clearer idea of the variables that influence the behavior during the start of the flow. The results obtained through the machine learning model will facilitate the analysis of the rheological data. In addition, further, understand the behavior of the gel force during the transition from solid to liquid. In addition to having a dynamic and adaptive model that determines the maximum pressure peaks during the flow start-up processes. The findings will also contribute to the understanding and modeling of non-Brownian suspensions in the viscoplastic matrix. This can be seen in the standard pumping processes of various industrial processes, especially well drilling in offshore operations. In addition, it can open discussion about the problems that originate in the measurement of this type of suspension.*

Keywords*: Machine Learning, Non-Newtonian fluid, Non-Brownian, Curve-fitting, flow start-ups.*

1. INTRODUCTION

The extrusion of systems containing a high-volume fraction of solids embedded in a liquid matrix is a common processing operation. It plays an essential role across numerous industries, with interests ranging from polymer (Rueda et al., 2017) to ceramic (Blackburn & Wilson, 2008; Powell et al., 2013) and from dental (O'Neill et al., 2017) to food applications, including an emerging interest in additive layer manufacturing (Kern et al., 2018). These highly concentrated suspensions, often referred to as pastes, show a complex flow profile where slip, shear, and plug flow regions co-exist (Ovarlez et al., 2009). Suspensions are mixtures of two or more substances that do not dissolve into each other and form new particles or compounds. Although these materials are commonly found in our daily life, such as juices, whole milk, and salad dressing (Sato & Cunha, 2009), or complex industrial processes, such as types of cement in civil engineering (Jarny et al., 2005) and drilling fluids in the petroleum industry (Andrade et al., 2018; Mendes et al., 2017), their mechanical properties are not easily obtained. Inaccurate rheological properties or misguided interpretation of the results can hinder process designs and make projects unfeasible. The rheological characterization of suspensions has been proven quite challenging. In the case of non-colloidal suspensions (particle radius ≥ 10 μm), different particle interactions (hydrodynamic, collisional, or frictional) may significantly affect the supposedly simple rheological flows. Depending on size, form, and concentration, particle distribution during rheological experiments may vary with shear stress and time, resulting in heterogeneous suspensions and misleading measurements. Wall slip (Koos et al., 2012), sedimentation, agglomeration, and particle motion are also factoring that influence the measurements.

In the case of simple shear flow between parallel plates, the disturbance created by an off-center particle interacts nonsymmetrically with the boundaries leading to the migration of the particle to the unique equilibrium position at the center. In the pressure-driven Poiseuille flow between parallel plates, the interaction of the disturbance created by the particle with the shear gradient of the quadratic flow generates a force directed away from the centerline. In contrast, the boundary interaction generates a force toward the centerline. These two opposing forces balance at a certain distance from the centerline, resulting in two equivalent equilibrium positions on either side of the centerline.
Predicting the rheological properties of suspensions like cement slurry or well abandonment fluids remains a significant challenge. Some complicating factors in modeling such a system include the size and shape distribution of the particles with cement, sand, and coarse aggregates. Another challenge is that most rheometers must be modified to accommodate large particles. These modifications lead to rheometers with geometries that do not allow for an analytical solution to the flow pattern, making it extremely difficult to determine rheological parameters such as yield stress and viscosity. During the development of a model, several considerations need to be considered: particle shape, spread size distribution particle, and the time evolution of the particle shape due to the interaction between the particles and the medium.

ML (Machine Learning) is a discipline that straddles the boundaries of computer science, statistics, artificial intelligence, and mathematical optimization. The structure of statistical and differential equation-based models is determined by assumptions about the system to be modeled. The strategy in ML is to let the data govern the model. Rather than establishing assumptions about the model's shape, an algorithm is used to grow up the model structure (Hansson et al., 2016). ML has different techniques such as NN (Neural Network), support vector machines, ANFIS (Adaptive Neuro Fuzzy Inference System), RT (Random Forest), and DT (Decision Tree) that show a good performance for prediction and classification problems (Mohaghegh, 2000). ML is widely applied in different engineering disciplines (Elsafi, 2014).
NNs are a subset of ML techniques that create a computational data-driven framework to reconcile the intricate relation between inputs and outputs. This is achieved by fitting the variables of each neuron to reduce the deviations between the actual and predicted data. Traditional NN training processes are carried out solely on a statistical basis. Over the years, different types of NNs were introduced, namely those of ANN (Artificial Neural Network), DNN (Deep Neural Network), CNN's (Convolutional Neural Network), and RNNs (Recurrent Neural Network). Each of these NNs has proven to be effective for various physical applications.

ANNs are information processing systems that are trained by using existing input/output data for obtaining the relationships between complex and non-linear input/output relationships. The studies about the usage of ANN in petroleum engineering show that ANN performs better against conventional approaches to various problems. Statistical tests can be used to validate the predictive model, making it possible to check the model's reliability. It is a non-parametric technique that makes no assumptions about the training data. It reflects human decision-making more closely than other techniques. The use of neural networks to parametrize general non-linear functions is well known, with several authors showing that is possible to approximate any continuous functions by these methods. NN also were successfully applied to predict the rheology parameters of specific, well behaved, mixtures (Franke et al., 2010; Gowida et al., 2019). This work aims to predict the start-up flow of non-Brownian suspensions in the non-Newtonian fluid by ML through frequent measurements of shear rate. The proposed models using DNN demonstrated good accuracy for various Carbopol predictions.

2. MATERIALS AND METHODS

2.1 Materials and Experiements

For the preparation of the physical gel, a dispersion of Carbopol Ultrex 10-20 was used at a concentration of 0.2% wt, neutralized with aminopropanol at 95% wt, also known as AMP95. The solution was mixed for 12 hours during neutralization, obtaining a Ph of 7.22 ± 0.1, and left to rest for a further 24 hours. The Carbopol solution concentration was determined with the objective that the yield stress not allow the sedimentation of the particles in suspension.

Carbopol suspensions were prepared with spherical polyamide glass particles with a density of 1150 Kg/m3. Particles of 20 μm, and 50 μm were used in concentrations of 1, 10, and 23 %wt, as shown in Table 1. To verify the particle size, we performed particle size distribution tests with Microtrac S3500. The particle size distribution tests showed that for particles of 20 μm [Fig.1(a)], more than 50% of the particles had this size, and for particles of 50 μm [Fig.1(b)], more than 60% coincided with the size of the supplier.

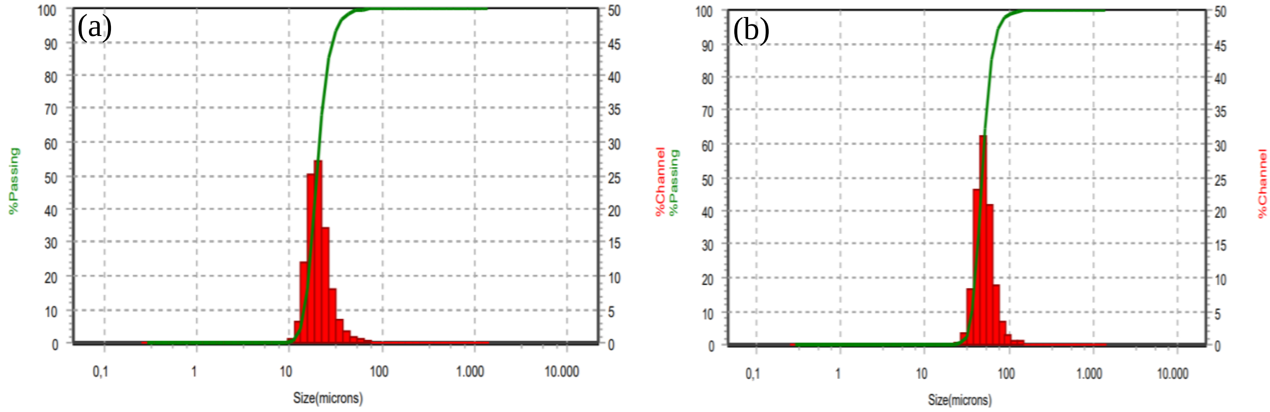

Figure 1. Particle size distribution (a) 20 μm size particle, and (b) 50 μm size particle.

Carbopol suspensions were tested in a rotational rheometer, DHR-3 (TA Instruments, USA), which is capable of measuring a minimum torque of 10^{8} N.m and has a maximum certainty lower than ±5% of the reading (Longo & Zilio, 2011). All experiments were performed at a temperature of 22°C and homogenized by an industrial mixer (Hamilton BeachHMD200) for 5 minutes at 1000 rpm to guarantee repeatability of the methodology. A 40mm diameter parallel plate geometry with a 1 mm gap and serrated surfaces to avoid wall slip (Dimitriou et al., 2011) was used to conduct all experiments. A sample hood was also employed to avoid sample drying.

Table 1. Different types of formulated Carbopol solutions with Polyamide seeding particles (PSP) are used for machine learning.

Sample Name	Particle size (μm)	Concentration wt%
Sample 1.1	20	0.5
Sample 1.2		5
Sample 1.3		15
Sample 2.1	50	0.5
Sample 2.2		5
Sample 2.3		15

Steady-state flow curves for Carbopol suspensions were conducted by imposing a series of constant shear rate levels ranging from 0.01 to 100 s^{-1} for 600 s each of the shear rates following the methodology proposed by Quitian et al., (2022), avoiding sedimentation.

Creep tests were developed to determine the behavior of the fluid before start-up flow. The test applied constant homogeneous shear stress from 5 to 75 Pa. Before each experiment, the fluids were pre-sheared at 30 s^{-1} over 60 s; after, the sample was kept left under a zero-shear stress condition for 300 s. Then, constant values of shear stress were imposed for 1000 s. When the shear rate drops monotonically approaching zero, the applied shear stress is considered to be below the yield stress, then when another stress is applied, and the shear rate tends to a steady-state value, then the applied shear stress is greater than the yield stress (Abedi et al., 2019; Barnes, 1999).

2.2 Prediction Models Development

This work used experimental creep data that were previously measured. Fig. 2 briefly shows the workflow followed to develop a robust ML model for rheological properties prediction. The workflow started with data acquisition and filtration to get high-quality data, the training stage for the machine learning model and optimizing the model hyperparameters, testing the model accuracy, re-training the model in case of low accuracy, and finally validating the proposed models.

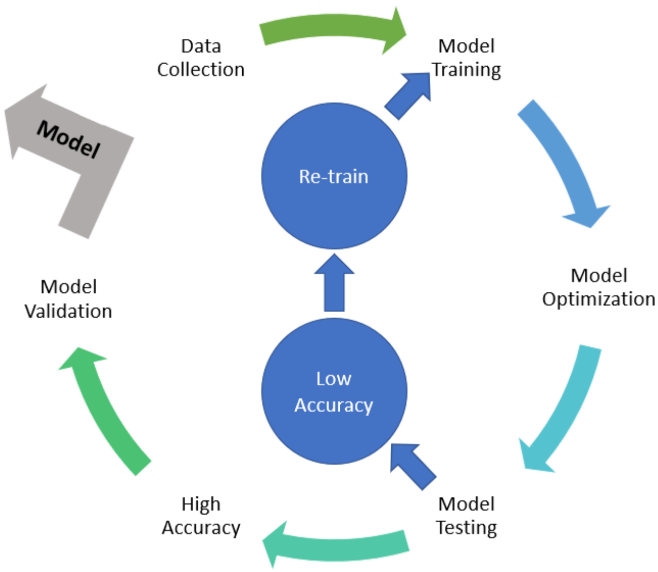

Figure 2. Flow chart for the start-up flow prediction model.

The data set used was collected and prepared to be used in machine learning processes. The data contains 180 data points of shear rate, shear strain, and time readings of different shear stress for each sample [Table1]. For avoided errors during the model's prediction, filtering and analysis of the data were performed to propose a better machine learning prediction. Finally, the statistical analysis of the data used for training and testing the developed models in this study.

2.2.1 Training NN

The NN training process is essential for learning the algorithm; here, the connection weights are the most appropriate (Rai et al., 2005). Many training algorithms start by assigning random numbers to the arrays of neurons. Simultaneously, the NN validates and fits the activation function. This is repeated until the validation error is acceptable. The machine learning process was performed through supervised learning. Supervised training consists of giving input and output data in advance of each experimental test (Aladjem, 1998). The training data goes through several epochs (iterations) until the experimental data matches the output of the NN. In this work, NNs architecture was employed to provide a predictive model for start-up flow of non-Brownian suspensions in the non-Newtonian fluid. The experimental data was divided into two groups: 80% for training and 20% for testing. The NNs hyperparameters should be tuned to get the optimum prediction performance.

2.2.2 Architecture of ANN

The NNs are built through several layers; the layers are organized into input, hidden, and output layers. Inside the layers, we have different neurons or perceptrons that connect to each other, and these connections go through the hidden layers to the output layer. The number of neurons in each hidden layer is an essential part of deciding the overall architecture of the NNs. The neurons are responsible for processing the data, thus influencing the final result. Using too few neurons in the hidden layers will result in something called underfitting (Rai et al., 2005). Underfitting occurs when there are too few neurons in the hidden layers to adequately detect the signals in a complex data set. Using too many neurons in the hidden layers can result in several problems. First, too many neurons in the hidden layers may result in overfitting. Overfitting occurs when the NN has so much information processing capacity that the limited amount of information in the training set is insufficient to train all of the neurons in the hidden layers.

An ANN is a parameterized mapping from input to output, given as a superposition of simple functions. In this study, the fully connected deep feedforward function was used. The network used input vector z of length n_1 and a weight matrix W_1 of size $n_2 \times n_1$ and a bias vector b_1 of length n_2 are introduced. Function f is a function of one variable and is called an activation function. A typical choice for this function is a sigmoidal function.

$$y_i = f\left(b_i + \sum_{j=1}^{n_1} W_{ij} z_j\right), where\ i = 1, \dots, n_2 \tag{1}$$

$$y = f(W_z + b) \tag{2}$$

This is a non-linear process and corresponds to a one-layer neural network. This is repeated for the next hidden layers defined. A DNN is defined by taking a superposition of such transformations. In Equation 3, it is shown how the NN transformations occur for different layers.

$$y_1 = f(W_1 z + b_1), \qquad y_2 = f(W_2 z + b_2), \dots , \qquad y_n = f(W_n y_{n-1} + b_n) \tag{3}$$

As mentioned earlier, the inputs to the DNN in our study are the parameters rheological of the fluid, the imposed shear stress, shear strain, Bingham model fit parameters, and time. These parameters are then correlated to a single output, shear rate, using a number of layers and neurons. The number of layers and neurons was varied until finding the best prediction of the model. Variables in a NN can be learned by minimizing the loss function according to Equation 4, which indicates the difference between the experimental and predicted values.

$$MSE = \sum \left(y_{Exp.} - y_{Pred.}\right)^2 \tag{4}$$

In this study, the depth of the NNs was changed from one to five layers, and the width was changed from 5 to 150 neurons per layer. The NN was evaluated, and it was observed that the network behaves with less error for the NN of two and three layers with 10 to 30 neurons, producing better levels of precision, and overfitting of the data was avoided.

Many times, the NN presents correlation problems, which need methods to optimize the output of the data. Currently, there are several methods that help solve optimization problems. The loss function is optimized using a combination of Adam's optimizer with a learning rate of 0.001, while the hyperbolic tangent function is used as the activation function where appropriate. For this word, Adam's optimization was used, which included a TensorFlow package. Table 2 summarizes the hyperparameters used to create the NN for predicting the start-up flow.

Table. 2 Main parameters used for the construction of the deep neural network

Parameter	**Value**
Training algorithm optimizer	Adam
Transfer functions	Tanh; Relu
Cost function	MSE or Loss
Number hidden layers	1 - 3
Number of neurons per hidden layer	10 - 30
Epochs	1 - 1000
Learning rate	0.001
Bias	Initial weights in 0

3. RESULTS AND DISCUSSION

This study aims to predict the start-up flow for different non-Brownian suspensions of Carbopol without the need to perform many rheological experiments. The creep tests are very time-consuming tests since need to discover the shear stress value at which the fluid begins to flow. For that reason, an NN was developed and trained. The NN was trained with 70% of the data from each fluid type, and the remaining 30% was used to test the behavior prediction.

The performance and accuracy of the developed NN were verified by comparing the predicted shear rate values with the experimental shear rate values. The training and test data were used to fit the weights of the connections and validate the relationship between the input and output of the NN. This process was carried out to find the best configuration and test the parameters of the configured NN until finding the most appropriate number of neurons and layers, avoiding over-fitting the model. It is essential to characterize prepared fluids before performing creep experiments, verifying that the fluids have a characteristic behavior of viscoelastic, viscoelastic, or elastoviscoplastic fluids since these types of non-Newtonian matrices present yield stress.

Steady-state flow curves were developed to identify the dynamic yield stress at various shear rates. Figure 3 shows shear stress as a function of shear rate for the six fluids prepared. The curves display that the suspensions of Carbopol with glass particles had a non-Newtonian behavior of the shear-thinning type. The curves were fitted to the Herschel-Bulkley model to determine an approximation of the yield stresses, which served as the basis for developing and schooling the shear stress values applied in the creep tests.

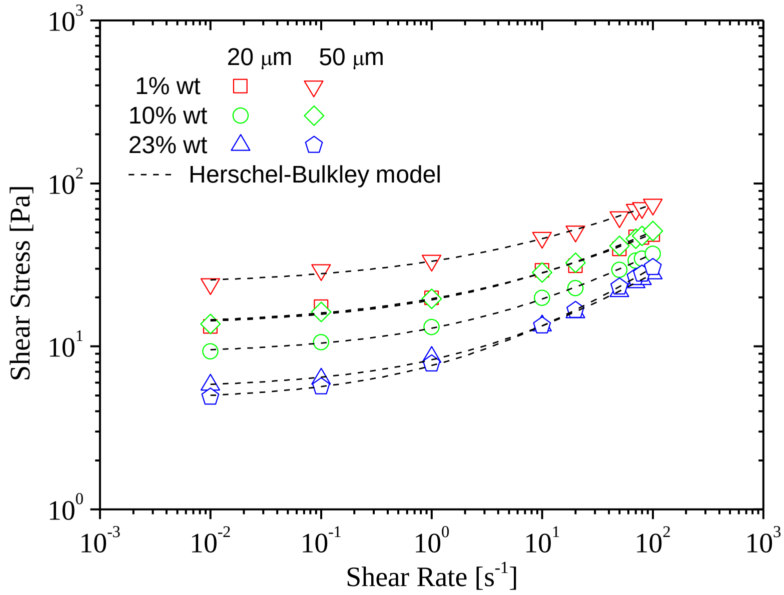

Figure 3. Steady-state flow curves at 22 °C were measured at atmosphere pressure on DHR-3 TA Instruments, with an applicating shear rate of 100 – 0.01 s^{-1}. Experimental data were fitted by the modified Herschel-Bulkley equation (dashed line).

The fitted parameters for each concentration are presented in Table 3. It is observed that the fitted values of the yield stress decreased with the increase in the concentration of the particles, as well as the Consistency coefficient and the flow behavior index. This phenomenon was evidenced in the suspensions with particles of 50 μm.

Table. 3 Fitted parameters of the Herschel-Bulkley equation. Yield stress (τ_0), Consistency coefficient (m), flow behavior index (n), and correlation coefficient (R^2)

Sample Name	$\tau_0[Pa]$	m [Pa.s^n]	n [-]	R^2
Sample 1.1	13.52	6.09	0.38	0.9866
Sample 1.2	8.99	3.95	0.42	0.9989
Sample 1.3	5.51	2.76	0.45	0.9996
Sample 2.1	23.80	9.52	0.36	0.9963
Sample 2.2	13.41	5.87	0.40	0.9991
Sample 2.3	4.67	2.95	0.47	0.9998

In training the NN, it is essential to decide which shear stress values will be used for training and testing. For this, it was necessary first to observe the behavior of the fluid. Observing the shear stress values where the fluid started-up its flow and did not flow, the shear stress values were chosen for training and testing in each suspension.

The results of the data chosen for testing are shown to know the behavior that the NN predicted. Figure 4 shows the evolution of the shear rate as a function of time for nine different values of shear stress. In Figure 4 (a), we have the yield stress values between 55 and 50 Pa, where a suspension of Carbopol with glass particles of 20 μm is necessary to start-up flow. It is observed that with the increase in the concentration of particulate, the fluid tends to become more brittle since the microstructure decreases, thus decreasing the yield stress values for the range of 10 to 15 Pa for a concentration of 10%wt. [Fig. 4(b)] and for a concentration of 23% wt. 5 to 10 Pa [Fig. 4(c)].

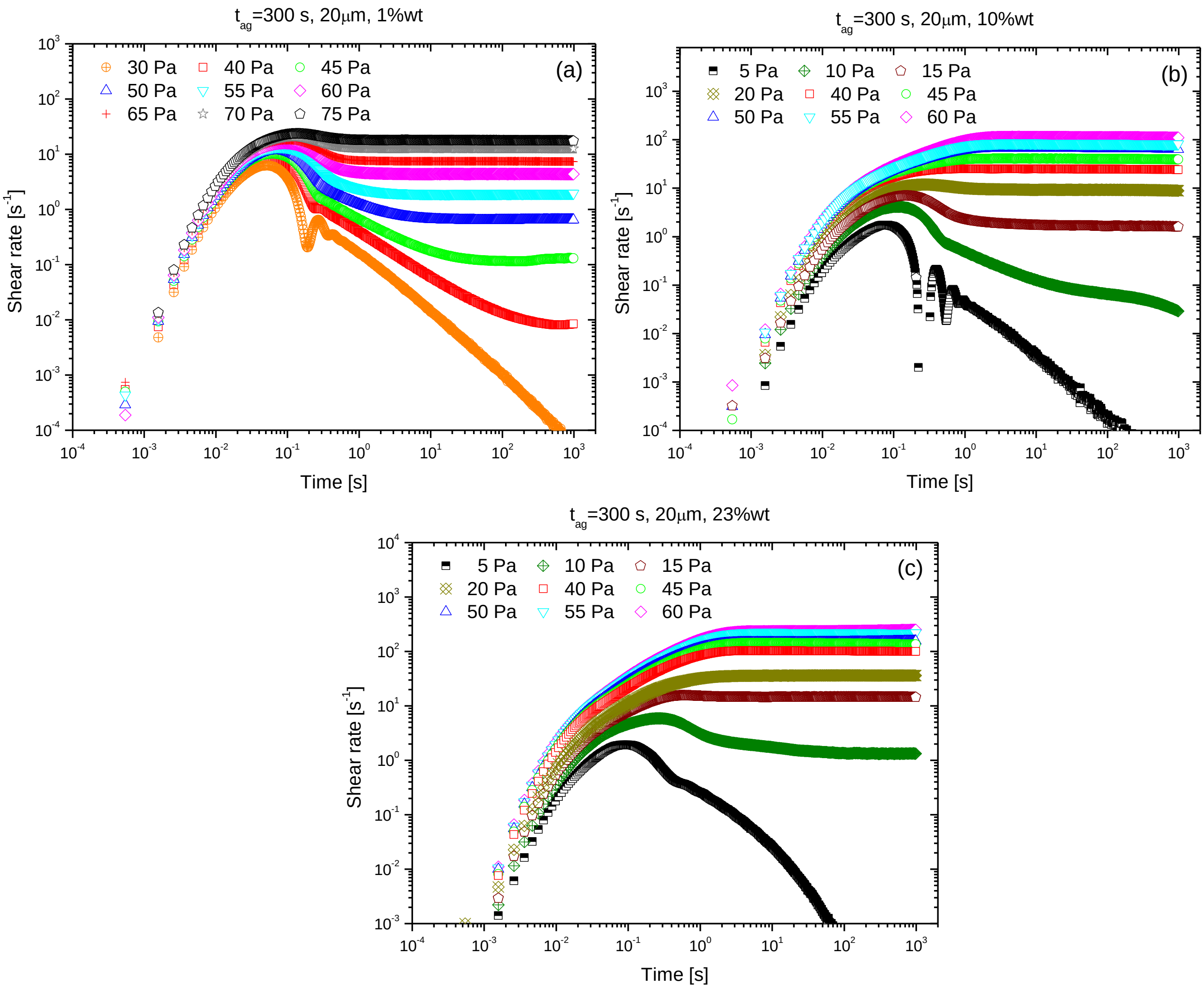

Figure 4. Shear rate as a function of time for different constant shear stress values of Carbopol suspensions with particles of size 20 μm and different concentrations (a) 1 %wt., (b) 10 %wt., and (c) 23 % wt.

Test data for fluids with 50 μm particles showed behavior similar to that of 20 μm, only with a more significant decrease in yield stress. For the standard fluid with a concentration of 1%wt [Fig. 5 (a)], the start-up flow shear stress range is between 40 to 30 Pa for a 10%wt. fluid [Fig. 5 (b)], the yield stress values decreased for a range of 20 to 15 Pa, and for a concentration of 23%wt of particles [Fig. 5 (c)], the yield stress must be in values very close to 5 Pa. By looking at the fluid start-up flow behavior for different particle types and concentrations, it is possible to determine that the same NN will predict the same behavior for these fluids.

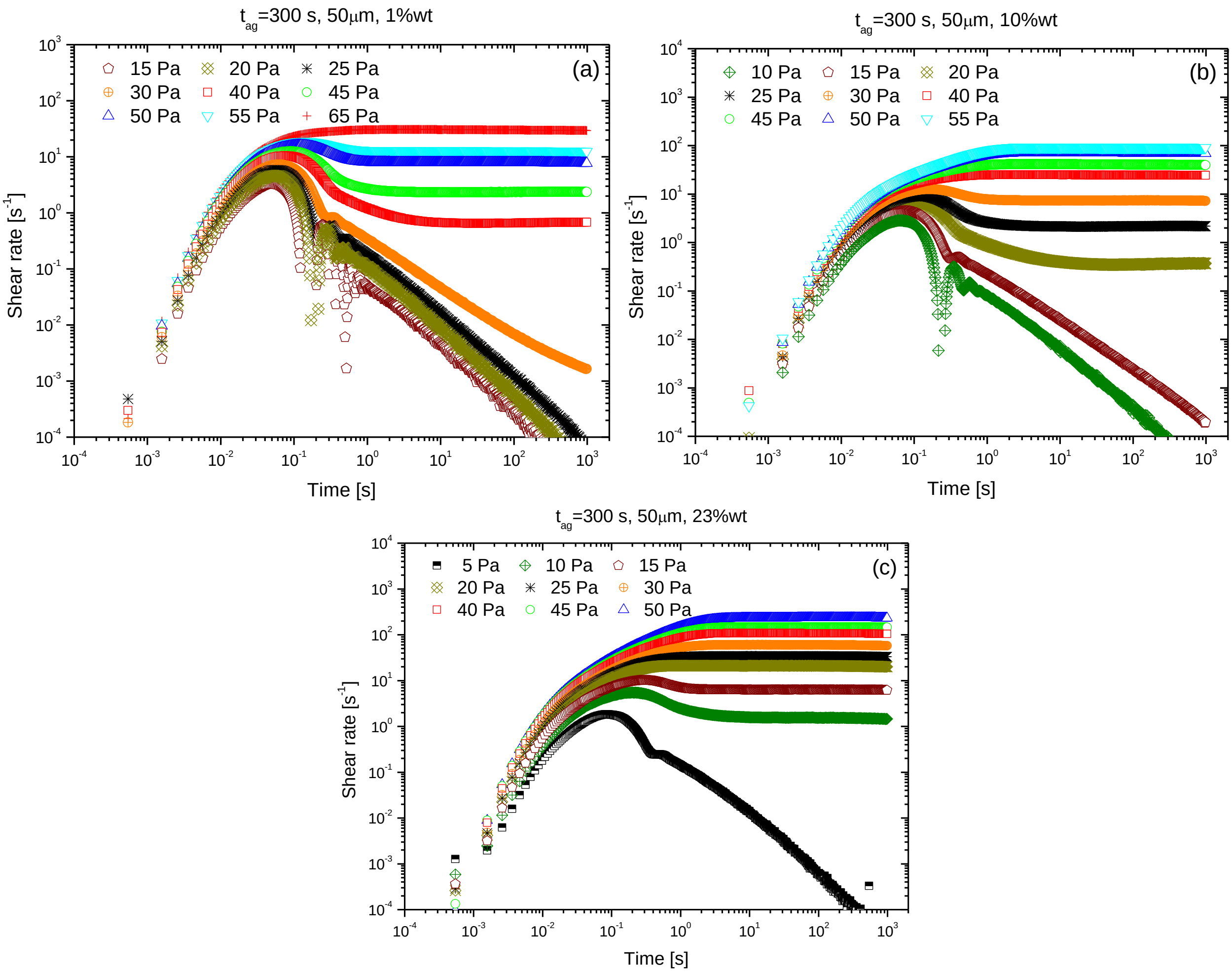

Figure 5. Shear rate as a function of time for different constant shear stress values of Carbopol suspensions with particles of size 50 μm and different concentrations (a) 1 %wt., (b) 10 %wt., and (c) 23 % wt.

It should be noted that the results predicted by the NN do not contain any physical intuition and are simply predictions based on experimental data of a specific composition of a fluid. This work aims to start a process of correlation of experimental data with mathematical and numerical models to determine the behavior of fluids with the same characteristics without the need to carry out many experimental tests.

In this study, during the data preprocessing, the data were normalized to facilitate the analysis and construction of the NN. As a result of this comparison, the most straightforward and precise model, the one that best fitted the experimental results, was selected for the rheological data set. As Figure 6 shows, there is an excellent correlation between the predicted and immediate solutions in this case, which suggests that the training is done correctly. The performance of the NN trained with experimental values predicted start-up flow values showed an average percentage error of 22.40% with a standard deviation of 27.85%. Figures 6 (a), (b), and (c) show the high agreement between the measured and predicted shear rate values of the developed ANN model, as shown in the cross plots.

The high precision of the developed model can be confirmed by the high R-value of 0.95 for training and 0.92 for testing, in addition to the low MSE of 2.8%, 4.5%, and 2.6% for the processes. Training and testing, respectively. This indicates that the ANN model was able to learn the relationships between the input and output variables. For given data sets. On the other hand, the suspensions with particles of 50 μm, presented fewer errors, 1.6%, 3.5%, and 2.2%. In Figure 7, it can be seen that the similarity of the predicted values and the experimental values coincided in the largest of the data.

Although selecting the number of layers and neurons is still trial and error, the process could be optimized to find the best values. These values of layers and neurons allowed us to determine when the data presents an overshoot and ends up not flowing. Its exact representation of that damping process would require models that are capable of predicting this behavior, as in the study of Andrade et al. (2016). It is essential to highlight that the network learning process was successfully achieved since the tested values were interspersed with the training values. This work is the beginning of developing an algorithm capable of mixing models with physical significance and experimental data.

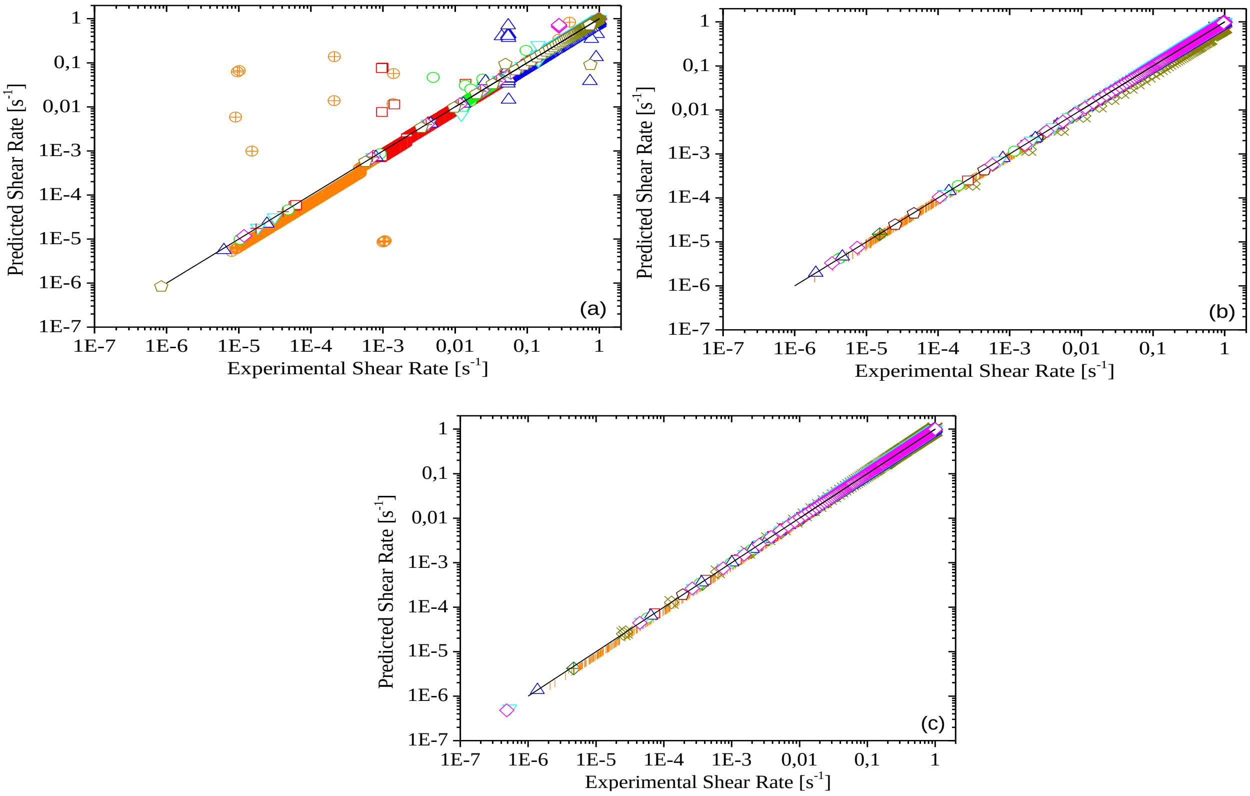

Figure 6. Comparison between predicted shear rate using ANN model with experimental data of Carbopol suspensions with particles of size 20 μm and different concentrations (a) 1 %wt., (b) 10 %wt., and (c) 23 % wt.

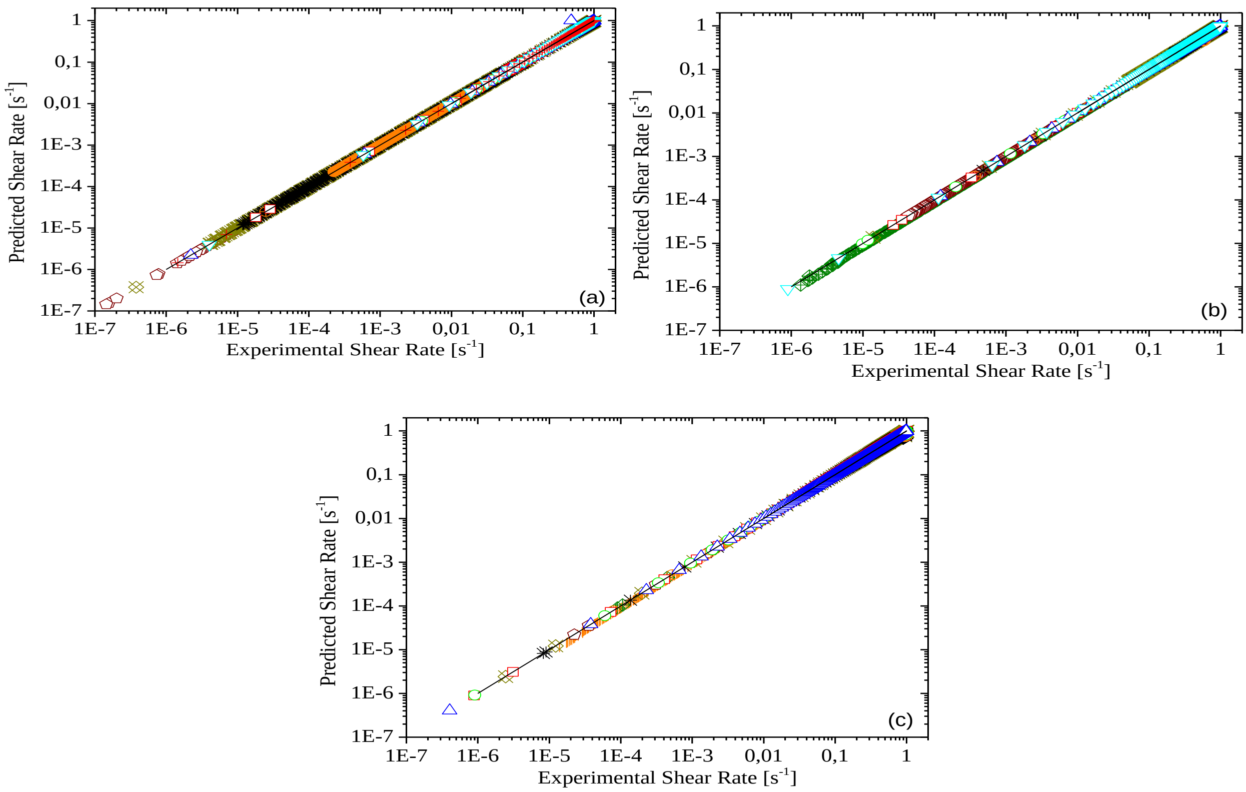

Figure 7. Comparison between predicted shear rate using ANN model with experimental data of Carbopol suspensions with particles of size 50 μm and different concentrations (a) 1 %wt., (b) 10 %wt., and (c) 23 % wt.

4. CONCLUSIONS

This study deals with predicting flux restart of non-Brownian suspensions using ANN. A multilayer feed-forward perceptron neural network was developed with the training algorithm to predict the shear rate as an output variable. The hyperbolic tangent functions were used as the transfer function in the hidden layer and the Relu function in the output layer. The optimization procedure selected the best ANN architecture to predict the target. Statistical analyzes showed that the ANN predictions were in remarkable agreement with the experimental data. In future work, it is proposed to seek to predict the behavior for a new formulation by extrapolating the measurements through models with physical significance. ANN can predict the rheological parameters with high accuracy r-value was more significant than 0.90, and AAPE was less than 6%

5. ACKNOWLEDGEMENTS

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