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Intelligent modeling and optimization of emulsion aggregation method for producing green printing ink

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Abstract: In this study, Artificial intelligence method was used as a new approach in modelling and optimization of printing toners with appropriate requirements. Toner fine powder is made up of resin, colorant and additives. This composite has been utilized in electrophotographic digital printing. The optimization approach has been considered for optimizing of toner production process and to produce printing toners with an appropriate physical and color properties (particle size (PS), particle size distribution (PSD), L*, a*, b*) by an environmental friendly method which is emulsion aggregation (EA). The EA is a green technology that provides many advantages for toner production pathway and lead to high quality product and printing. The effect of heating rate (R), time of mixing (T), and mixing rate (S) on PS, PSD, and L*, a*, b* has been studied. An in-home code was established to optimize the architecture of artificial neural network (ANN) with two hidden layers, by which an accurate model was developed for the prediction of toner properties. The best process conditions with acceptable characteristics of manufacturing toners was obtained by multi-objective optimization in specified amounts of heating rate, mixing time, and mixing rate.

Keywords: environmentally toner; emulsion aggregation; digital printing; artificial neural network; NSGA II

1 Introduction

Toner is a composite of resin, colorant, special agents for charge controlling and other essential additives for the printing. This composite has wide application as an ink in electrophotographic printing process [1-3]. Generally, printing toners are categorized into two main classes; first class of toners is dry powder type that this type of toners has a particle size (PS) in range of 5-10 µm. On the other hand, the second class of toners is liquid dispersed type with a PS around 0.1-3 nm. It is worth denoting that, dry powder type has many advantages over liquid dispersed type in synthesis and application point of view. Moreover, dry powder type toners has widespread application for printers and copying machines in home and office [4,5]. Extensive application of these materials makes it vital to consider effect of them on environment when designing toners. In the same field, the European Computer Manufacturers Association (ECMA-328, 2007) has hinted the significance of checking and recording of particle discharge to the ambient from laser printers by preparation a standard [6]. Therefore, green process for toner production widely has been considered in recent years [4,7].

Physical properties of toner particles are very important for digital printing quality. This parameter can change the digital printing characteristics of laser printers and photocopiers. It is worth denoting that, majority of toners physical properties are dependent on synthesis method and well-established methods can lead to favored homogeneity, particle size (PS), particle size distribution (PSD) and shape of toner particles in final product [8]. However, ordinary approaches normally based on grinding were founded to be unsuitable to produce small particles with narrow size distribution and spherical shape [9]. Therefore, new production method or developing of traditional products was taken into consideration by researchers in academic and industrial projects [10].

In addition to noted physical properties, toner microstructure has a great effect on printing quality. Modern techniques for toner production have

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several advantages over the traditional methods. For an example, in situ polymerization can lead to excellent situations of toner in size, shape, and size distribution. Although, polymerization processes for toner manufacturing has environmental limitations [11]. Recently, emulsion aggregation method (EA) as a new process has been developed for toner manufacturing. This technique is a chemically controlled method with ability of achievement to small particle size, uniform shape and favored particle size distribution. However, main advantage of this process is being environmentalfriendly based on toner production process and final product [12].

In EA process, toner production precursors has been mixed for a specified time at a given temperature in an aqueous medium for toner particle formation [13]. In this method, required energy for toner production and printing of them in consequent applications has been fallen. A detailed report from Xerox Corporation Ltd. showed the aforesaid advantages of EA approach. Therefore, EA process has been considered as an environmentally friendly method compared with all other toner production routes [14,15].

Based on a series of prior publications, Diverse black printing toners have already been manufactured via suspension [8], emulsion aggregation [9], emulsion, and mini-emulsion [10] procedures with an entirely a large variation of physical and color qualities. Toners with different nature were also prepared by changing the kind and amount of coloring agent [11,12,16]. Though the modeling/optimization technique used in our recent paper was successful in detecting and optimizing properties of toner, and RSM and DF can properly find the best solution for one or binary response, but this feature is suffering from the quality of optimization of all responses together and even in the overlaid plots which should be in the special level of factors one cannot find the optimized area. Normally, one may apply classical or stochastic modeling approaches for thorough identification of behavior of a given system [17,18]. The use of artificial intelligence-based models instead of polynomial-based ones provides with an opportunity for the mathematical tool to learn the behavior of the process, to generalize the behavior learnt to thousands of experimental cases not examined yet, and to make a decision for selecting the best set of input variables needed for achieving predetermined properties [19]. Therefore, to optimize all responses together we try artificial neural network (ANN) in the same content level also by ANN one finds factors for special responses. Authors are specially focused on producing toner and

optimizing the condition to find the best solution to produce toner with selected specification.

In the current work, we applied, for the first time, artificial intelligence approach to detect and identify variation pattern of toner properties in terms of processing variables. We wrote an in-house computer code based on ANN that makes possible intelligent modeling and optimization of printing toner properties. It is worth denoting that, the ANN modelling approach was used for process modelling and a multi-objective optimization method based on genetic algorithm was utilized for process optimization.

2 Experimental

2.1 Materials

The polymer used was a styrene-acrylic resin (NS88; Simab Resin Co., Iran). A polyethylene emulsion wax (EE 95, Kala Kar Co., Iran) and a carbon black pigment (Printex U, Degussa-Evonik, Germany) were also used. Polyaluminum chloride and nitric acid (Merck Co.) was used as coagulation agent. Deionized water and 4% NaOH (Merck Co.) solution was used as medium and neutralization respectively.

2.2 Toner production procedure

A stepwise method was used for toner production based on previous reports [16-18,20]. First step (step a) of this method was preparation of base suspension in a 1-liter beaker by 3 g wax, 2 g carbon black, 24.5 g styrene-acrylic latex, and 120 g deionized water. Solution preparation process was finished by manually mixing for about 15 min at room temperature. Second step (step b) of base solution preparation was of suspension by a homogenizer (5 min). Then, the mixture was stirred for 1 h to achieve a homogeny suspension (step c). In the following, by the accession of a solution of 0.6 g coagulation agent in nitric acid over 10 min, the PH value of solution was reached to 2. Main product of this process in this step was a gel with a clear change in viscoelastic behavior. Next step in toner production procedure (*step d*) was rising the temperature of the mixture to 50°C while the gel was continually stirred. This process was continued for about 30 min. As the step e, temperature of the mixture was increased to 96°C in a time step of 30 min and it was kept in this temperature for 60 min (step f). The last section of this pathway for toner production (step g) was holding the product of step for a future 60 min at 96°C.

Product mixture of noted process was neutralized with NaOH solution and was cooled down to reach the room temperature. Final product (toner) was achieved by some next post-processes; insolating from the water, washing to remove divalent ions, filtering, and drving.

2.3 Characterization

To check the printability of the toner, they were applied to a paper substrate using a monochrome laser-jet printer (HP 1100, Laser-jet printer).

Particle size (PS), particle size distribution (PSD) of the toner was measured on a particle size analyzer (PSA, Malvern Mastersizer 2000, England).

The color characteristics were estimated employing Ihara SpectroCam (Japan).

3 Model development and optimization

In this work, GA-based optimization was implemented to ANN models developed based on experimental data. In the first step of modeling with the artificial intelligence approach, we obtained ANN models. In the second step, models were applied in the optimization of toner properties on the basis of a reciprocal intelligent tool. Three parameters containing heating rate (R), mixing time (T), and mixing rate (S) were selected as input variables to assess their effect on particle size (PS), particle size distribution (PSD) and L*, a*, b* (output variables), individually and simultaneously. Accordingly, experiments were performed to obtain sufficient data to be fed into ANN models (Table 1). The training of ANN allows for learning the appropriate behavior of the selected property [22-25]. The weights and biases of the ANN are to be found in a manner to minimize prediction error made by the network in the course of training [26,27]. When the training has been completed, ANNs are able to foresee toner upon receiving any input similar to the pattern they are taught. According to Table 1, different scenarios were feeding into ANN models assigned to each property of printing toner. Hyperbolic tangent sigmoid activation function was employed to normalize input variables in [-1, +1] interval [20]:

$$X_{i} = 2 \times \left[\frac{x_{i} - x_{\min}}{x_{\min} - x_{\max}} \right] - 1 \tag{1}$$

where X_i denotes a normalized value of input variable x_i , while x_{min} is minimum and x_{max} is maximum value of target functions, respectively.

An in-house code was developed in this work that can conceptually search for best configurations, i.e., for

Table 1.	Scenarios	considered	as internal	data for	the constr	ucted model.
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Scenario	1 st input (x ₁) R (hr.)	2 nd input (x ₂) T (hr.)	3 rd input (x ₃) S (rpm)	1 st output (y ₁) PS (µm)	2 nd output (y ₂) PSD (-)	3 rd output (y ₃) <i>L*</i>	4 rd output (y ₄) a*	5ª output (y₅) b*
1	1.0	1.0	1000	5.43	2.22	9.49	-0.92	-0.71
2	0.5	1.0	500	4.99	1.80	11.55	-1.65	-0.45
3	1.0	1.0	1000	5.08	2.44	8.51	-2.43	-1.4
4	0.5	1.5	1000	8.72	1.97	14.21	-1.5	-0.52
5	1.5	0.5	1000	7.83	2.10	10.67	-1.87	0.79
6	0.5	0.5	1000	9.20	2.47	9.87	-1.45	-1.45
7	1.0	1.5	1500	5.37	1.59	11.97	-2.68	0.31
8	1.0	1.5	500	5.28	2.41	9.44	-2.58	0.02
9	1.0	0.5	1500	8.30	2.13	13.18	-1.72	-0.85
10	0.5	1.0	1500	0.96	1.11	8.04	-4.25	0.21
11	1.5	1.0	500	12.78	2.36	10.58	-2.31	0.29
12	1.0	1.0	1000	5.18	1.60	9.23	-1.8	-0.97
13	1.5	1.0	1500	1.11	1.29	11.63	-1.7	0.84
14	1.5	1.5	1000	6.07	2.02	8.31	-3.07	0.31
15	1.0	0.5	500	4.19	1.20	5.60	-2.66	0.01

dividing data into "training", "validation", and "test" sets. Accordingly, 75 percent of data were chosen to be fed to model for training. On the other hand, 15% of data were selected for validation of ANN model. Finally, other data were utilized for testing of configured ANN structure. Although, there is no rigid rule to find the appropriate number of neurons in the hidden layers, complexity of the relationship between inputs and outputs plays a key role [28,29]. Different combinations of neurons (one to ten neurons in each hidden laver) in two hidden lavers were tested for choosing the best network with minimized error. The function defined through Eq. 1 is applied in output and hidden layers were used as an activation transfer function:

$$f(x) = \frac{e^x - e^{-x}}{e^x + e^{-x}}$$
 (2)

According to the flow sheet shown in Figure 1, a wellorganized computer code was written to intelligently model properties of the toner. To optimize parameters of the model, i.e., to find the best weights and biases that

match the input and output variables, a gradient descent (GD) method. Number of neurons in hidden layer 1 (NH1) and hidden layer 2 (NH2) was varied in the range of 1-10 (maximum level of neurons in each hidden layer (max NH1 and max NH2) was considered equal to 10). The reliability of ANN model was measured by means of mean squared error (MSE) criterion:

$$MSE = \frac{1}{n} \sum_{i=1}^{n} \left(Y_i - \overline{Y}_i \right)^2$$
 (3)

where n is the number of samples, Y_i and \bar{Y}_i are experimental and predicted value of response for sample i.

In the light of the above procedure, one ANN model was yielded to separately predict of study effective parameters on printing toners. In the next step, for the multi-objective optimization of toner production process a Non-Dominated Sorting Genetic Algorithm-II (NSGA-II) was utilized. For the production of chromosomes, the value of each gene was randomly chosen in view of the values of variables given in

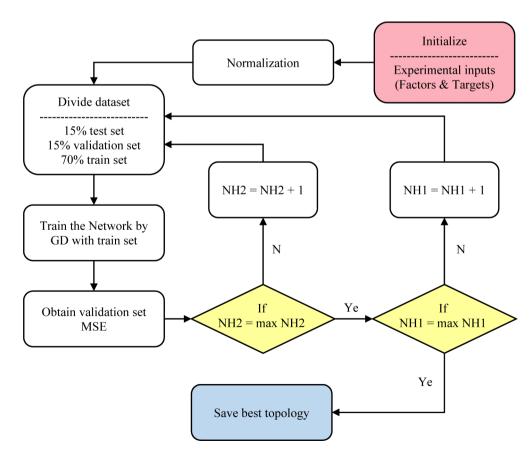


Figure 1: Modeling flowchart based on ANN method.

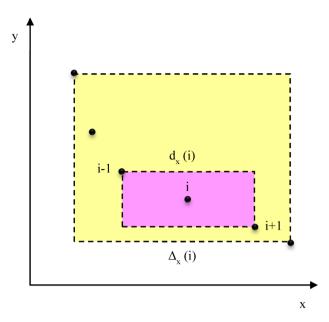


Figure 2: Crowding distance calculation parameters for sample.

Table 1. The code was then implemented to find fitness of each chromosome to simultaneously optimize toner properties. After giving pairs of chromosomes were compared together, they were isolated from the remainder, as the first Pareto front. This operation was surveyed to obtain Pareto fronts 2, 3, etc. Pareto front *n* was assigned to the chromosome that has been dominated (n-1) times. We also used crowding distance (C.D.) values, as the second criterion for optimization, was to sort Pareto fronts:

$$C.D.(i) = \sum_{x=1}^{N} \frac{d_{x}(i)}{\Delta_{x}(i)} \quad where: \quad d_{x}(i) = |F_{x}(i+1) - F_{x}(i-1)|$$

$$\& \quad \Delta_{x}(i) = |\max(F_{x}) - \min(F_{x})|$$
(4)

In the above equation, N is the number of objective functions, C.D. (i) is crowding distance of chromosome i, and d_{x} (i) and Δx (i) based on the objective function x is shown in Figure 2. After estimating the extent of fitness based on primary and secondary constraints, chromosomes were sorted only by the most favorable to unfavorable order. The best chromosomes were chosen as the first Pareto front, followed by selecting, pairing, reproducing, and muting. Parents and children of the new generation were revisited for fitness utilizing nondominated sorting algorithm. Table 2 gives the values of parameters employed for optimization of toner properties on the ground of NSGA-II evolutionary algorithm (Figure 3).

Table 2: The value of parameters used for multi-objective optimization by GA

Optimization parameter	Value		
Initial population size	50		
Crossover mechanism	SBX		
Crossover rate	70%		
Mutation mechanism	Gaussian		
Mutation rate	40%		
Selection mechanism	Ternary Tournament Selection		

4 Results and discussion

Normally. for toner production with suitable characteristics, PS, PSD and color characteristics should be controlled. In case of the EA process, to the best of the authors' knowledge, there is no report on the optimization of the parameters based on ANN. The effect of mixing rate (S), mixing time (T), and heating rate (R), used as parameters to determine PS, PSD, and L*,a*,b* of printing toner yielded from the green emulsion aggregation (EA) with ANN.

In this study, to unveil the complex nature of the relationship between inputs and outputs, an artificial neural network was used. The optimized structure of ANN with 8 neurons in each of hidden layer 1 and hidden layer 2 was achieved (Figure 4). Selecting of best ANN structure in this study was done by considering the validation step MSE. Between the different probable networks with 1 to 10 neurons in each hidden layer, a combination of 8 neurons in hidden layer 1 and 8 neurons in hidden layer 2 was introduced as the best structure. This network has a 3 level input (x1, x2, and x3) and 5 outputs (Y1, Y2, Y3, Y4, and Y5).

4.1 Modeling of toner production with ANN

The performance of the developed ANN model is checked by comparing the experimental and predicted values of each response. From Figure 5, errors in prediction of each response are really minor. As shown in this figure, some of the experiments are not fed into the ANN network for training and these experiments were used for the model accuracy study. Significant similarities between the ANN model outputs and experimental results show the good accuracy of model predictions. From the statistical point of view, the slope and intercept of the regression equations for the outputs are very close to 1 and 0, respectively.

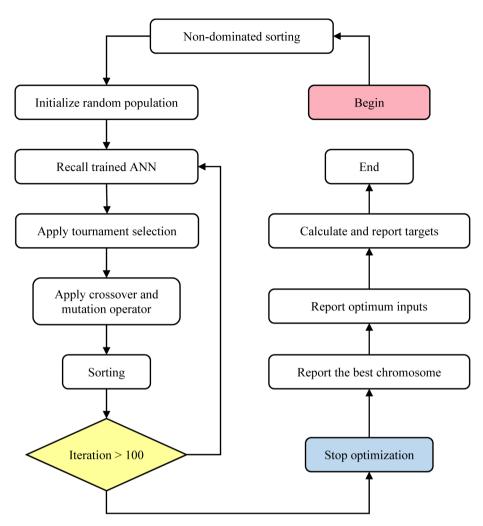


Figure 3: Flowchart of NSGA-II based optimization.

Furthermore, modeling accuracy based on R calculation was calculated about 0.99, 0.991, and 0.88 for training, validation, and testing section, respectively. Finally, R for all experimental data compared with modelling results was achieved higher than 0.96.

To get a brighter prospect of the accuracy of the optimized ANN developed in this work, the quantities of target functions for the whole trained, validated, and tested data sets are compared in Figure 5. The plots show that the optimized ANN predicts each response well. It is worth announcing that, Figure 5 could indicate the accuracy of modelling (R > 0.96). However, a clear comparison of experimental data and modelling results for each sample are shown in Figure 6.

Granting to the findings of ANN modeling in the present survey, the three-dimensional surface plots were used to depict the relationships between input and

output variables. Figures 7-9 shows three-dimensional (3D) plots of the relationships between the influence of R, T, and S on PS, PSD, and L*, a*, b* respectively derived from the above model. In each figure, the effect of two input factors was studied on the obtained PS, PSD, and L*, a*, b* respectively, and the values of the remaining input factors were fixed at low, medium and high-range value.

The formation and population of particles is mostly controlled by the physical chemistry of the mixture [30]. As illustrated in Figures 7-9 the simultaneous manipulation of heating rate and mixing time results in a significant change in the PS. It is worth denoting that, increasing the mixing time and along with it reduction the heating rate leads to a parabolic-like trend in particle size. On the other hand, simultaneous change in speed of agitation and heating rate will be lead to abrupt change in the toner PS. Based on

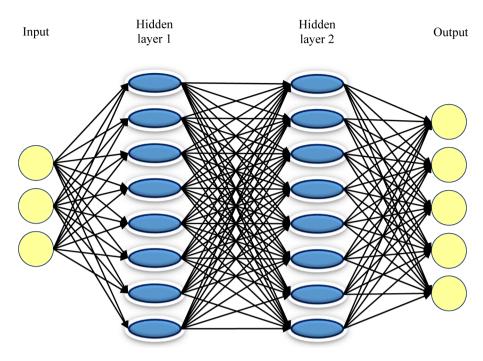


Figure 4: Optimized structure of ANN.

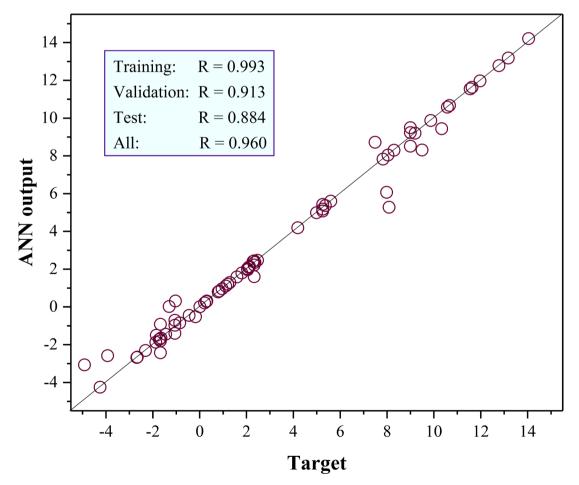


Figure 5: Relationship between the experimental data and the optimized ANN model outputs.

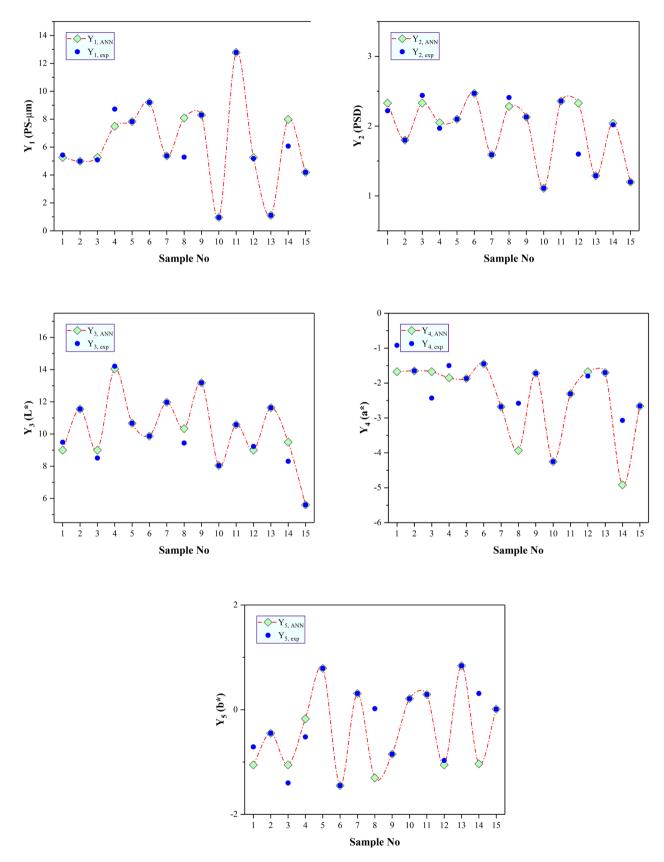


Figure 6: Comparison of the experimental with predicted values corresponding to PS, PSD, and color (L*, a*, b*).

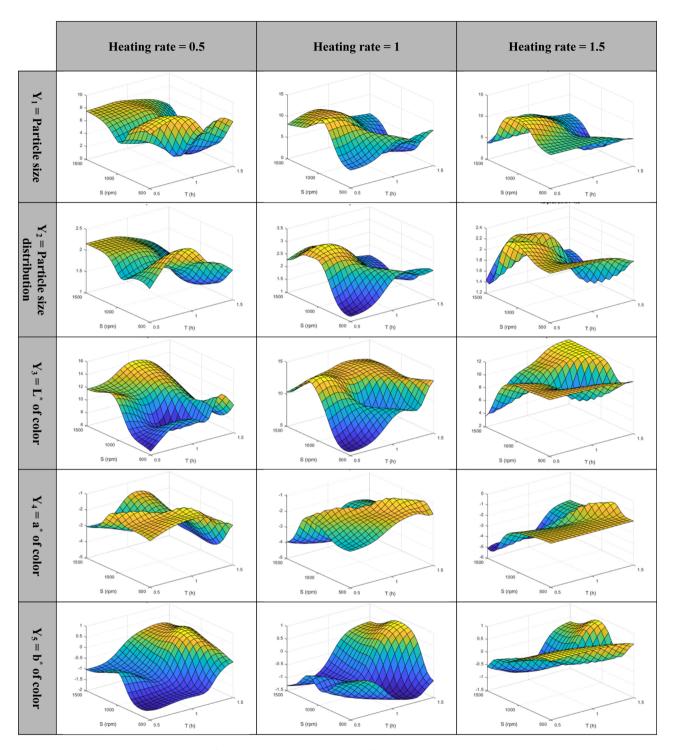


Figure 7: 3D plots of modelling responses (PS, and PSD, and color (L*, a*, b*)) in constant values of heating rate.

experimental results and comparison of them with ANN results, it seems that production of toner with suitable PS by prediction of production process conditions is accomplishable. A reduced PS upon growing of agitation speed could expected, in view of higher breakage rate being considered at raised speed of rotation. On the

other word, the final size of particles is depended on the balance between breakage and coalescence [30,31].

An approximately similar tendency was saw in L*(lightness), which can be ascribed to the reality that this color specification correlates fairly good with the PS. More evidence comes from the reality that the volume of

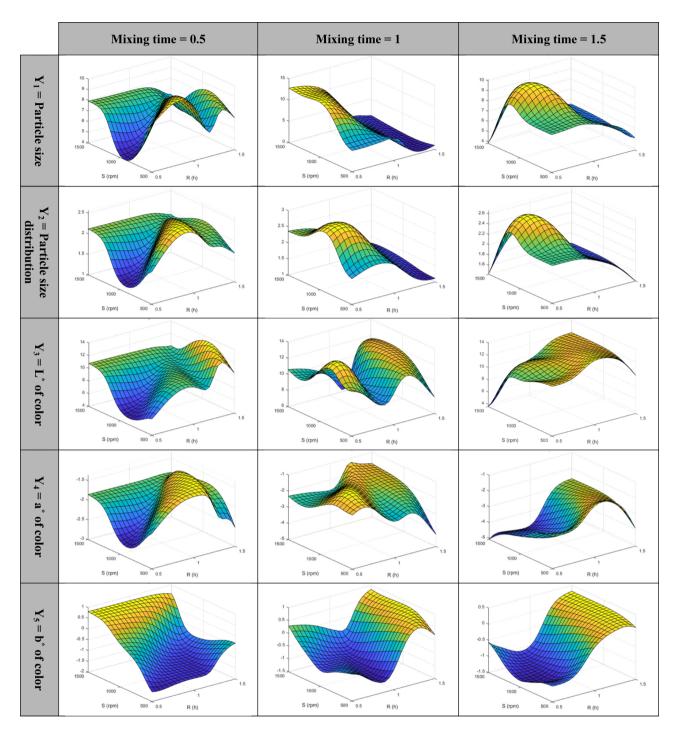


Figure 8: 3D plots of modelling responses (PS, and PSD, and color (L*, a*, b*)) in constant values of mixing time.

carbon black has hardly been reformed by the researchers, therefore, the L* of toner should be monotonically governed by the size of toner particles [3,8]. In this point of view, the incompetence of the utilized model to account for a* and b* can be rationalized. Actually, progress in aggregation, even after assumed period of process, will be controlled by the collision frequency between particles [21,30,32].

ANN is a mathematical pathway for process modelling. It should be denoted that, after process modelling, an optimization method should be applied on ANN model for calculation of the best conditions for achieving favorite responses [31]. Therefore, this study was continued by GA-based multi-objective optimization with NSGA II.

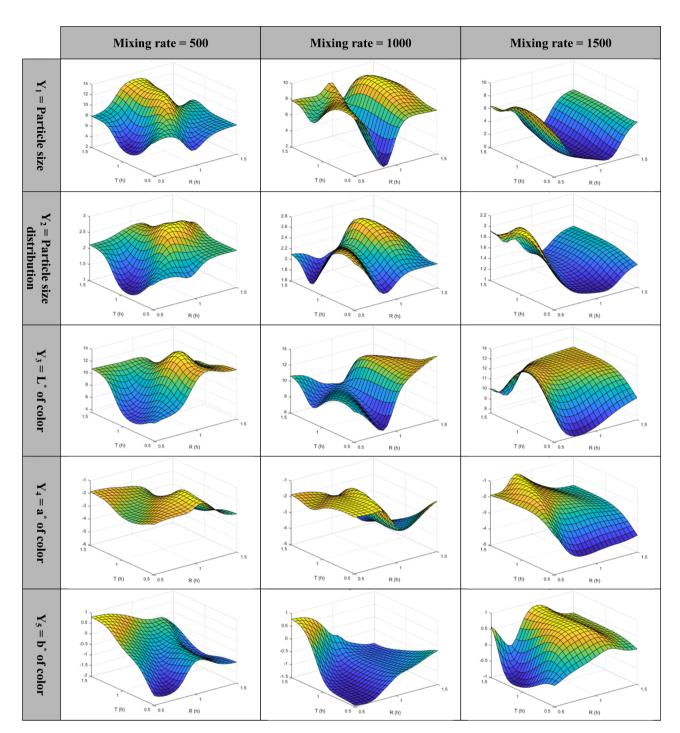


Figure 9: 3D plots of modelling responses (PS, and PSD, and color (L*, a*, b*)) in constant values of mixing rate.

4.2 Multi-objective optimization of toner production based on ANN and generic algorithm

In wide range of process, there is not a single point as the optimal solution with respect to all objectives. Therefore, a set of solutions are optimal points that each of them has on or more excellence over other solutions. This set of solutions have been known as Pareto optimal solution [21,32]. There are wide range of classical manners and evolutionary algorithms for solving the Multi-objective optimization (MOO) problems. MOO problems in classical methods have been solved by converting the MOO problem to a single

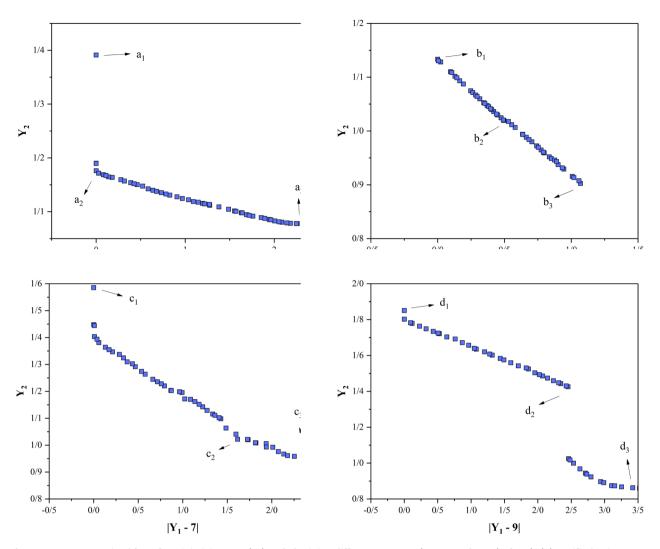


Figure 10: Pareto optimal front for minimizing PSD (Y2) and obtaining different amounts (3, 5, 7, and 9 μm) of PS (Y1) (specified points are introduced in supporting data).

objective optimization problem. In the other pathway of classical methods for MOO solving, MOO problem will be solved based on one object. In the following, other constraints will be applied on achieved solutions and the best solutions will be chosen. weighted sum method, e-constraint and goal programming are some of the classical methods for MOO solving [32]. Changing the MOO problems to single objective problem, usually leads to much easier solving and less computational cost. However, this method is not efficient for wide range of problems [21]. On the other hand, there are several evolutionary algorithms to solve MOO problems such as genetic algorithm. Solving the Multi-Objective problems by Genetic Algorithm (MOGA) was introduced by Fonseca and Fleming firstly in 1993. There is a major difference between the MOGA and standard GA that is in ranking GA population (MOGA uses the non-dominated classification concept in ranking of GA population) [21,33]. It is clear that, MOGA can be used for any mathematic models to solve a multi-objective problem (of course, with a little change in circumstances). In this study, a trained ANN was used as a mathematical model fed to evolutionary algorithm for optimizing the toner production process conditions.

Succeeding to developing of the models, an effort was made to optimize toner production (heating rate (R), mixing time (T), and mixing rate (S)) for a desired particle size (3, 5, 7 and 9), particle size distribution (minimize), L*, a*, b (37, 0, 0). These objects were searched by different amounts of inputs in specified range fed to validated ANN model. First multi-objective optimization was done for minimizing the PSD and PS reach to specified values (3, 5, 7 and 9). The Pareto optimal set for these objects is presented in Figure 10. Plotting the corresponding

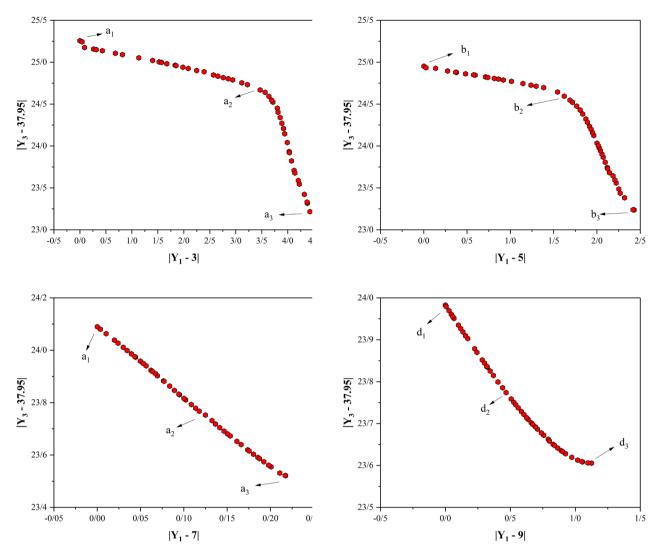


Figure 11: Pareto optimal front for obtaining L* (Y3) equal to 37.95 and different amounts (3, 5, 7, and 9 μ m) of PS (Y1) (specified points are introduced in supporting data).

decision variable against one of the objectives makes the understanding of their role in generating the optimal Pareto set possible.

As results given in Table 1, this procedure of green toner production can produce toners with wide range of characteristics. For an example, toners with PS in range of 5 to 12 μ m were produced. Normally, a unique PS of about 12 μ m ends in a print resolution of 300 dots/inch (dpi). Less diameter toners of around 8-10 μ m are necessary to have a print with enhanced resolution (about 600 dpi). However, grinding toners with larger diameter than 12 μ m is not appropriate and leads to dots and ragged lines in printed images and copy quality will be degraded [30]. It was also understood that best conditions required to find the minimum possible PS and PSD are not basically the alike for color characteristics [31,33]. In general, the results are indicative of the

importance of multiple optimization technique for finetuning of the toner characteristics.

In addition to Y2, Y1 is an important response of process that can be changed by Y1 variation. In other word, specifying an object for Y1 can change the amount of Y3. Results of multi-objective optimization of Y1 and Y3 are shown in Figure 11. As noted previously, Y1 in range 3-10 can considered as optimum level. However, 3, 5, 7, and 9 were considered as the numerical optimums of Y1 and optimum level of about 37 was specified for Y3. Four Pareto optimal fronts for noted conditions were achieved and are shown in Figure 11. In addition, minimizing of Y5 is optimum level for this parameter. As presented in Figure 12, multi-objective optimization of Y5 and Y1 was done. It is clearly visible that, instead of previous optimizations, these objects are accessible in large number of points.

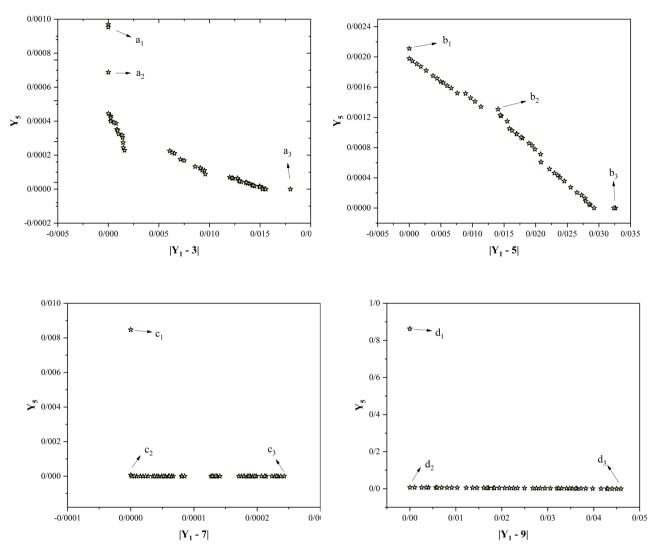


Figure 12: Pareto optimal front for minimizing b* (Y5) and obtaining different amounts (3, 5, 7, and 9 μm) of PS (Y1) (specified points are introduced in supporting data).

As the final MOO, Y2 and Y5 minimizing, Y4 and Y5 minimizing, as well as Y2 minimizing and Y3 about 37 were investigated. As shown in Figure 13, several points for these objects are reported. a* and b* minimizing is favorable for toner because, this conditions shows that color quality was remained fixed in process duration. On the other hand, PSD should be very low for a high quality toner. It seen in Figure 13 that many points with very low PSD are accessible.

5 Conclusion

A green rout for toner production based on emulsion aggregation is presented in this report. Particle size and size distribution controlling for achievement to toner

particles with small size and narrow size distribution is necessary for accessing to a suitable toner with high quality image on substrate. However, this process is a multi-objective problem and needs detailed modelling and optimization procedure for specifying the optimum conditions. A multi-layer perceptron artificial neural network modelling and response surface methodology was used for process modelling and NSGA II was applied on ANN model for optimization of EA process variables (mixing time, mixing intensity, and heating rate) and achieve to suitable predict about physical and color characteristics of toners. In both RSM and ANN methods, good fitness (with R higher than 95%) was achieved and it was also found that increasing the time or agitation rate does not only guarantee the production of smaller toner particle with acceptable size distribution and colorimetric

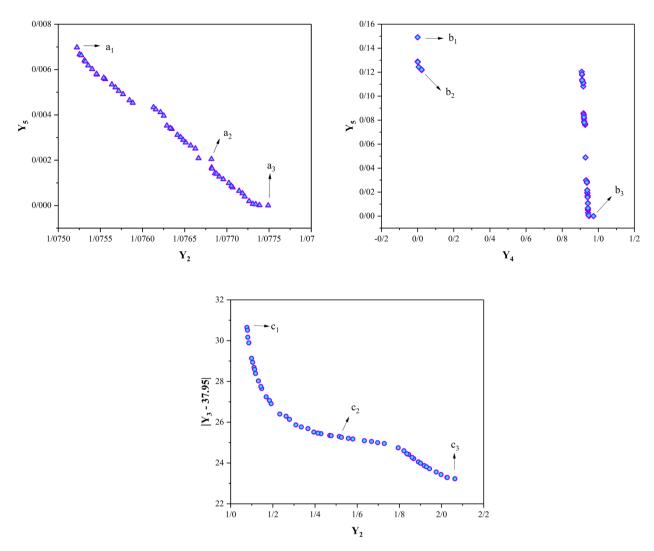


Figure 13: Pareto optimal front for (a) minimizing PSD (Y2) and b* (Y5), (b) minimizing a* (Y4) and b* (Y5), and (c) minimizing PSD (Y2) and obtaining L* (Y3) about 37.98.

assets. The multi-objective optimization was done to estimate optimal conditions for production of a toner with PS of 3, 5, 7, and 9 µm. On the other hand, minimum values of PSD lead to the best quality toner. Moreover, each of other objects has unique optimum level that should be considered in NSGA II. Finally, several points are reported to achieve these objectives.

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