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Neural network simulation for non-MSMPR crystallization

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Abstract

A neural network model has been developed for the simulation of steady state industrial crystallizers where, in general, the crystal size distribution cannot be described by simple mass and energy balances, i.e. they are non-MSMPR crystallizers. The model is based on fundamental equations of steady state suspension crystallization. The parameters in the nucleation rate have been chosen for the simulation of different chemicals. The particle size distribution of the product is expressed by the Rosin–Rammler equation. Different operating modes and deviations in crystal size distribution caused by the suspension being imperfectly mixed are presented by different values of modified Rosin–Rammler number. The ranges of variables in the neural network have been chosen based on data for industrial crystallizers. The dominant size of particle, and the productivity of the crystallizer can be predicted with input information. Thus, this neural network can be used for most chemicals and for different kinds of operating conditions. The results predicted with the neural network have been verified by solving the fundamental equations and by comparison with experimental data.

Keywords: Crystallization; Neural network; Simulation; Design

1. Introduction

Fast simulation of a process is very useful for process optimization and control and the neural network is a powerful tool for fast simulation of complex processes. After the neural network has been trained, the training program is no longer needed. Only the weight file and the short neural network code are required to simulate the process. It is easy to transfer the neural network code to other programs to present the results as figures. The neural network can respond to the input variables very fast and gives simulation results over a wide range of variables. Industrial crystallization includes mass and heat transfer with complicated nucleation and growth processes and is affected by many factors, mainly kinetic parameters, mixing conditions and operating conditions. Simulating and controlling the crystallization process in an industrial crystallizer becomes very complicated. Many simulation programs have been developed for various crystallization processes [1,2]. However only a few works have tried to simulate the crystallization process using a neural network for the control of a distributed parameter crystal growth process [3]. The aim of this work is to develop a neural network for simulation of a continuous process in a non-ideal industrial crystallizer so that the result can be used as a basic tool for optimizing and controlling continuous crystallization.

2. Theory of the neural network used

For decades, much progress has been made in understanding the crystallization process. In a continuous crystallization process, when the system is operated under steady state, the basic properties of the particle and solution, the basic parameters in the crystal growth rate and nucleation rate, and basic operation conditions all affect the particle size distribution in the product. For a description of the solid product, the production rate and the particle size distribution are important. There are many mathematical expressions that can be used to describe the particle size distribution [1]. Toyokura [4] has used the Rosin–Rammler equation to describe the particle size distribution in the product. In the Rosin–Rammler equation the modified uniformity number, m, and the dominant size of particles have been used to express the particle size distribution. The value of the modified uniformity number, m, is dependent on the operation mode of the crystallizer. In the ideal case the uniformity number is unity, and then the equations become the conventional ones, e.g. those studied by Randolph and Larson [1] in their study of MSMPR (mixed suspension mixed product removal) crystallizers. There are also inter-

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Nomenclature

B	nucleation rate (#/m^3 s)
C_0	feed concentration (kg/m^3)
C^*	equilibrium concentration (kg/m^3)
ΔC	supersaturation (kg/m^3)
D	impeller diameter (m)
G	crystal growth rate (m/s)
i	the power of the crystal growth rate in nucleation rate model (–)
j	the power of the suspension density in nucleation rate model (–)
k	the power of the impeller tip speed in nucleation rate model (–)
K	product of the crystal density and volume shape factor (–)
K_N	constant in nucleation rate model (–)
K_v	volume shape factor of crystal (–)
L_d	dominant size of crystals (m)
M_T	suspension density (kg/m^3)
N	stirrer speed (1/min)
p	productivity (kg/m^3 s)
P	production rate (kg/s)
V	volume of crystallizer (m^3)
V_0	feed rate (m^3/s)
V_t	impeller tip speed (m/s)
V_w	evaporation rate (m^3/s)

Greek letters

ε	volume fraction of solution (–)
ρ_c	crystal density (kg/m^3)
τ	residence time (s)

actions between the crystal growth rate, nucleation rate and operation conditions in the crystallization process.

In this work, a neural network is developed based on the Toyokura theory [4], which has been proved useful by many industrial crystallizer data and has been used for the successful design of many industrial crystallizers. The theory and the basic equations are as follows.

A continuous crystallizer is supposed to be operated under steady state conditions and the cumulative size distribution of the product crystal is assumed to be expressed by the Rosin–Rammler equation on population basis as

$$r = \exp\left(-\frac{L}{L^*}\right)^m = \exp(-x^m) \tag{1}$$

where r is the relative cumulative number of product crystals, m the modified uniformity number, which is a constant particular for each product (it is independent of particle size), L the size of crystal, L^* the particle size corresponding to r of 0.3679, which is independent of the value of the modified uniformity number, x the dimensionless size of crystal $x = L/L^*$.

Based on this assumption, the production rate, crystallizer volume, nucleation rate, growth rate, and the parameters of the Rosin–Rammler equation are related. The relationship has been based on the population balance, and mass balance as given by Toyokura and Sakai [4]:

$$\frac{P}{\rho_c V} = L^{*3} B k_v m \int_0^\infty x^{m+2} \exp(-x^m)\,\mathrm{d}x \tag{2}$$

$$\frac{PL^* \int_0^\infty x^m \exp(-x^m)\,\mathrm{d}x \int_0^\infty x^3 \exp(-x^m)\,\mathrm{d}x}{\rho_c V \int_0^\infty \exp(-x^m)\,\mathrm{d}x \int_0^\infty x^{m+2} \exp(-x^m)\,\mathrm{d}x} = (1-\varepsilon)G \tag{3}$$

The modified Rosin–Rammler number, m, and particle size L^* present the shape of the particle size distribution in the product. The modified Rosin–Rammler number, m, depends on the operation mode, for example fines destruction and classified product removal, on the hydrodynamics in the crystallizer, and on the geometry of the crystallizer. When the system of crystallization operation is fixed, the value of the modified Rosin–Rammler number, m, is independent of operation conditions. The value of the Rosin–Rammler number can only be obtained by knowing the particle size distribution under the operation mode. The nucleation rate, B, is affected by supersaturation of the solution, the suspension density in the crystallizer and the mixing intensity. The nucleation rate also depends on the scale of operation, geometry and hydrodynamics of the process. If the nucleation rate is controlled by secondary nucleation, it can be expressed by the following equation.

$$B = K_N G^i M_T^j [f(N, D)]^k \tag{4}$$

As many authors have reported [5–8], the nucleation rate can be scaled-up based on the mixing intensity. By using different expressions for the mixing intensity, such as specified power input or constant tip speed of the impeller, the power law model of the nucleation rate can be used in different scales of crystallizers.

The crystal growth rate is mainly affected by supersaturation of the solution. It can be expressed by the following equation:

$$G = K_g (\Delta C)^g \tag{5}$$

Together, population balance, mass balance, with nucleation and growth rates give a perfect description of the crystallization.

3. Architecture of the neural network

The purpose of this work is to develop a neural network to correlate basic system information and operation conditions to the particle size distribution and to other important information about the crystallization process. The growth

rate and nucleation rate under steady state can also be obtained. As introduced above, the particle size distribution is expressed by Rosin–Rammler equation. Dominant particle size and the modified Rosin–Rammler number should be taken into the neural network. Whether or not a crystallization system operates under ideal mixing, with the operation mode fixed, the modified Rosin–Rammler number is constant. It can only be obtained by experiment. Therefore it was considered as a system parameter and used in the input layer. Dominant particle size and productivity are important quantities in a crystallization process and were chosen as outputs. On the other hand, nucleation and growth rates depend on the supersaturation in the crystallizer. For steady state crystallization the supersaturation level depends on the interaction between the crystal growth rate and nucleation rate. It cannot be determined from the operation conditions. Therefore, the nucleation and growth rates are also important data for the analysis of the crystallization processes. Therefore, these two parameters were also chosen as outputs, making four outputs altogether. The following quantities were chosen to serve as inputs:

- the operation conditions, which are feed concentration, feed rate, equilibrium concentration which corresponds to the crystallization temperature, mixing intensity, suspension density and the volume of the crystallizer,
- the modified Rosin–Rammler number, which is the operation mode,
- the properties which characterize the chemical, for example, particle properties,
- the parameters of the nucleation rate.

The parameters in the nucleation rate depend on the properties of the particle, the geometry and scale of the crystallizer. The nucleation rate model should describe the behavior of nucleation for different scales of a crystallizer. The criterion for the scaling-up of the nucleation rate was based on the mixing intensity used. With this method, the effect of the scale of the crystallizer on the particle size distribution is present. The operation conditions and the properties of the particle and the parameters of the nucleation rate together decide the dominant size of the particles. Basic mass balance determines the productivity. As introduced above, the Rosin–Rammler equation is used to present the particle size distribution in the product. The modified Rosin–Rammler number, *m*, is an important parameter in describing the operation mode. For example, if the operation is with classified product removal, the particle size distribution becomes narrow whereas the value of *m* becomes large. It means that the width of the distribution is presented by *m*. By knowing the value of *m* and the dominant size of particles the particle size distribution is predicted. The hydrodynamics of the crystallization affects the modified Rosin–Rammler number, *m*, in a way that determines what kind of classification is produced. We only know the average value of the Rosin–Rammler exponent as obtained from an operating plant. The hydrodynamics is different in different locations in the crystallizer tank. The present method cannot give detailed information about the effect of hydrodynamics on the crystallization. For that purpose a more fundamental approach is needed which is outside the scope of the present work.

Considering the basic equations in the theory section, it can be seen that, in order to simulate the crystallization, the properties of the product, basic information about the solution and the solid properties, the kinetics parameters, the operation conditions, and the operation mode parameter must all be known. For a certain continuous crystallization process, the properties of the solution and particle, such as the density of the particle, and the parameters of the nucleation rate are constant. Therefore, only the operation conditions, such as the residence time, feed concentration, mixing intensity, and the parameter *m* are needed to obtain the properties of the product. Here, we have a system of equations that is difficult to solve analytically. Moreover, in general, the parameters of the nucleation rate are not known. In this case, the use of a neural network is much simpler. A very simple neural network, different from the one presented here, would take information out of an operating industrial crystallizer and would build a network based on these data. The network could then be used for simulation and optimization. This is the conventional use of a neural network in an industrial process. But such a network would work only for that single equipment and for a single chemical whereas in this work we try to develop a neural network which can be used for most systems used in industrial crystallization. Therefore, the parameters in the nucleation rate were included in the neural network of this work.

The architecture of a multi-input and multi-output neural network is shown in Fig. 1. Input of the neural network is at two stages, one is the neural network layer and the other is the user layer. The actual input layer of the neural network is calculated from the user layer. The two input layers are used for the following reasons. The user layer can be used as neural network input, but the input layer in Fig. 1 has fewer inputs which results in the network being easier to

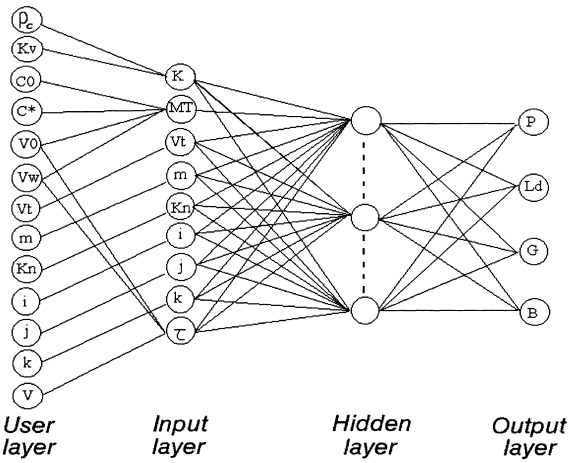

Fig. 1. The architecture of a neural network for simulation of crystallization.

train. The input layer can also be used such that the user has to calculate some inputs himself. The architecture of the network shown in Fig. 1 is easy to use and such a network is easy to train. There are 13 inputs in the user input layer, nine inputs in the neural network input layer and four outputs in the output layer.

4. Training and results

Training the neural network is an important step for developing a useful neural network. In order to have a range of variables that are valid in most industrial systems, the range of the neural network inputs and outputs were defined as follows:

Stirrer tip speed: 1.5–4.0 m/s,
Rosin–Rammler number: 1.0–3.0,
Suspension density: 20–320 kg/m^3,
Product of the particle density and shape factor: 520–4420,
Residence time: 1000–6000 s.

Parameters in the nucleation rate model:

$$B = K_N G^i M_T^j v_t^k \quad (6)$$

where $K_N = 1.2 \times 10^7$–9.6×10^{24}, $i = 1.0$–2.5, $j = 0.8$–1.5, $k = 1.0$–3.7.

The range of the output:

Productivity: 4.09×10^{-3}–0.1 kg/m^3 s,
Dominant size: 1.03×10^{-5}–1.82×10^{-3} m,
Growth rate: 1×10^{-9}–1×10^{-7} m/s,
Nucleation rate: 2.07×10^3–1×10^{10} #/m^3 s.

The data for training and testing the neural network are produced by solving the basic equations of the Toyokura design theory, Eqs. (1)–(5). 1500 sets of data were produced and used to train the neural network. The result of rough training to find the number of suitable hidden nodes are shown in Fig. 2. From this figure it can be seen that the average error of the training data and test data is very small when the number of hidden nodes is larger than 14.

Based on the results shown in Fig. 2, 16 hidden nodes were selected for accurate training of the neural network.

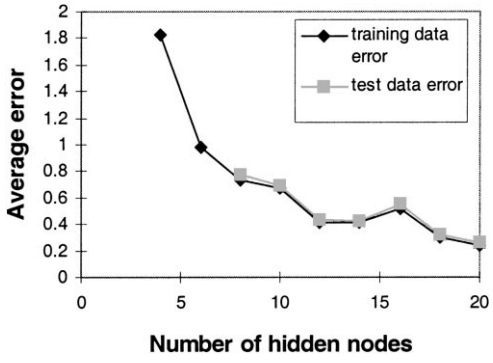

Fig. 2. The average errors of the training data and test data defined as relative mean deviations.

The results of accurate training were that the average error as relative mean deviation for all outputs was 0.097% and the mean correlation coefficient was 0.9994. Another 1500 sets of data were produced and used to test the neural network. The relative mean deviation for the test data was 0.100%. It is clear that the neural network describes the crystallization system very well.

5. Verification of the neural network

The purpose of the verification is to show that the neural network can produce the same results as the theoretical model. The results should also be similar to experimentally obtained data. This part of the work is presented in Section 6.

The simulated results of the relationships between the inputs and outputs of the neural network have been verified by solution of the basic equations. The results predicted by the neural network were compared with the results obtained from solution of the fundamental equations. The necessary test inputs for the neural network were chosen randomly within the interval used for training the network; they were not used in the training of the neural network. The accuracy of the neural network was expressed by the relative mean deviation.

Typical results for the effect of residence time on the productivity, dominant size of particle, nucleation rate and growth rate are shown in Figs. 3–6. The input data used for the neural network and fundamental equations are as follows:

Stirrer tip speed: 2.196 m/s,
Rosin–Rammler number: 1.578,
Suspension density: 196.3 kg/m^3,
Residence time: 1000–6000 s,
Product of the particle density and shape factor: 3907.

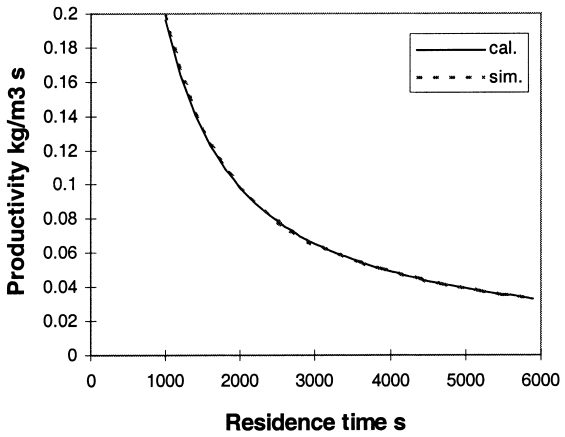

Fig. 3. The relationship between the residence time and productivity. “Cal” refers to the results obtained by solution of the fundamental equations. “Sim” refers to the results obtained with the neural network simulation. All figures use this same notation.

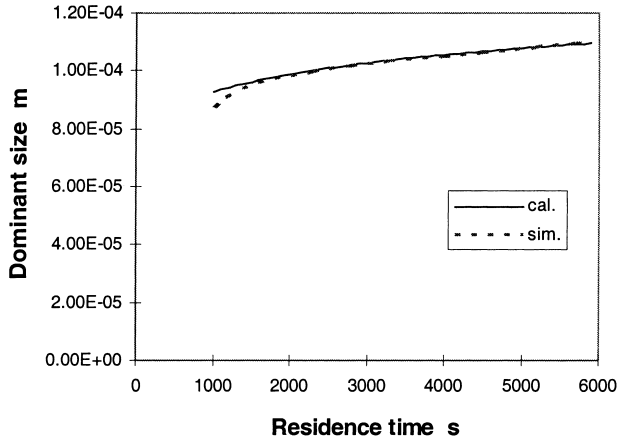

Fig. 4. The relationship between residence time and dominant size.

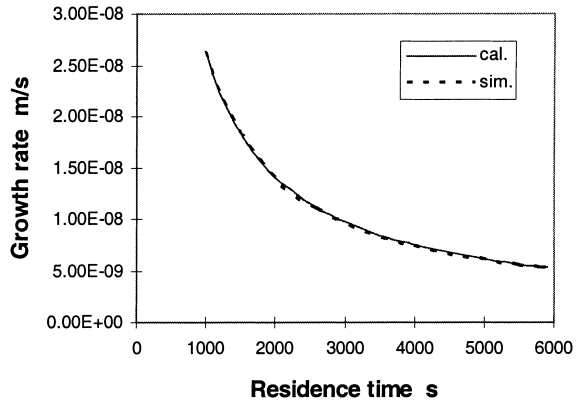

Fig. 5. The relationship between residence time and growth rate.

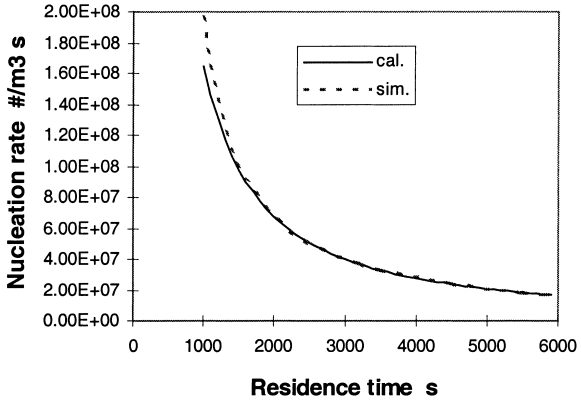

Fig. 6. The relationship between residence time and nucleation rate.

Parameters in the nucleation rate model (by Eq. (6)):

$$B = K_N G^i M_T^j v_t^k$$

where $K_N = 1.2\times10^{15}$, $i = 1.421$, $j = 1.194$ and $k = 1.31$.

The average training errors as relative mean deviations in percent for each output are:

Productivity: 0.29; dominant size: 0.722,
Growth rate: 0.464; nucleation rate: 1.63.

For the relationship between the residence time and productivity as shown in Fig. 3 the neural network works very well. There are nearly no differences between the values predicted with the neural network and those from fundamental equations. The productivity decreases with increasing residence time because the feed rate, feed concentration and crystallization temperature are constant.

The simulated results of the neural network for the relationship between the residence time and dominant size is shown in Fig. 4. The error appears in the range where the residence time is smaller than 1800 s. However, the average error as relative mean deviation is still very small, 0.722%. The crystal size is affected mainly by the growth rate and residence time. The increase in dominant size caused by increasing residence time is shown in this figure.

The relationship between the residence time and crystal growth rate are shown in Fig. 5. An increase in residence time will result in a decrease in the supersaturation in the mother liquor. Consequently, the growth rate decreases. The effect of residence time on the crystal growth rate is clearly shown in this figure.

The results obtained with the neural network and by solution of fundamental equations for the relationship between residence time and nucleation rate are shown in Fig. 6. It shows that the nucleation rate was simulated very well by the neural network when the residence time is larger than 1500 s.

6. Verification with experimental data

As shown in Section 5, the neural network can produce the same results as the solution of the basic theoretical equations. It means that the model of the neural network is correct. In this part of the work, we are going to verify the neural network model with experimental data. The work will be done with the following method. The basic operation conditions will be the same as those for the experiment. The parameters in the nucleation rate were obtained with experimental results. Then the neural network simulated the steady state nucleation rate. It was compared with the data obtained experimentally under steady state. In this way it can be proved that the neural network can produce reliable results. For this purpose, the results produced by the neural network developed in this work were verified by experimental data presented by Qian et al. [8,9]. They measured the crystal growth rate and nucleation rate in different sizes of crystallizers for potassium chloride in water solution. The nucleation rate and growth rate can be expressed as

$$B = 4.65 \times 10^{22} G^{2.78} M_T^{1.21} V_t^{3.79} \quad (7)$$

$$G = 2.32 \times 10^{-5} \left(\frac{\Delta C}{C^*}\right)^{0.91} \quad (8)$$

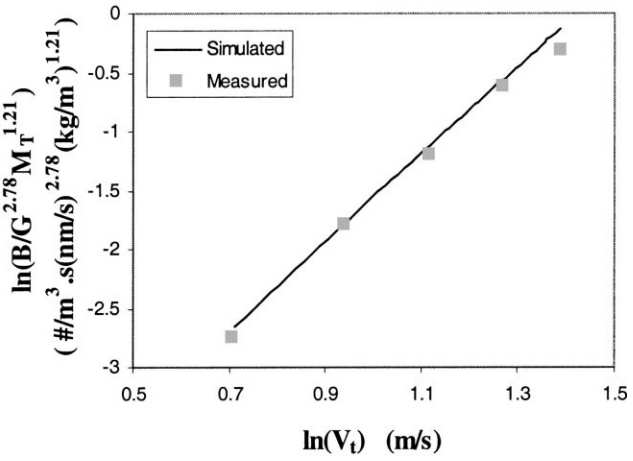

Fig. 7. Comparison between the simulated results and experimental data presented by Qian et al. [8] for the relationship between the stirrer tip speed and the parameters of crystallization kinetics.

The relationship between crystallization kinetics and impeller tip speed, V_t, was measured for different operation conditions. The measured data presented by Qian et al. and simulated results with the neural network have been shown in Fig. 7. From the figure it can be seen that the simulated results fit well to the experimental ones. This means that having the parameters in the nucleation rate equation available, the neural network can produce results similar to experimental data.

It is to be noted that we have verified firstly that the neural network produces the same results as solution of the system theoretical equations and secondly that the neural network produces the same nucleation rates as experimental data. We have not carried out verification of all the output variables against experimental data. However, the theoretical equations we are using here have shown that the neural network is verified experimentally.

7. The method of use of the neural network

The neural network simulator needs information about the basic properties of the chemicals, for example particle density and shape factor, and the kinetic parameters. The kinetic parameters should be obtained experimentally. The kinetic parameters should come from a crystallizer similar in geometry to the real crystallizer used. In this case, the crystal size distribution is available from the same experiments from which the kinetic data have been obtained. The Rosin–Rammler number, *m*, will easily be obtained from the particle size distribution. When the basic input information is available, the neural network can simulate the crystallization process when the values defining the basic operation conditions are varied as shown in the section describing verification. The neural network gives the results of productivity and dominant particle size. The crystal size distribution can be calculated with the Rosin–Rammler number, *m*, and the dominant crystal size. This information is usually a requirement for a crystalline product in an industrial process.

This neural network can also be used to determine the volume of crystallizer and the stirrer speed for a known geometry of the crystallizer. The method for designing a crystallizer is as follows. The specified dominant size of product and the production rate are required. The neural network input related to the crystallizer volume is the residence time. Therefore, the relationship between the residence time, productivity and the particle size will be found by the neural network. When the input range of the residence time is sufficient, the dominant crystal size is found as the output. The corresponding productivity is also read. From the residence time and productivity, the product rate for unit volume of crystallizer is calculated. Therefore, the crystallizer volume is calculated by the production rate of unit volume of the crystallizer. Different mixing intensities are used to find out a suitable impeller tip speed. The design procedure is shown in Fig. 8. The reliability of the design of the crys-

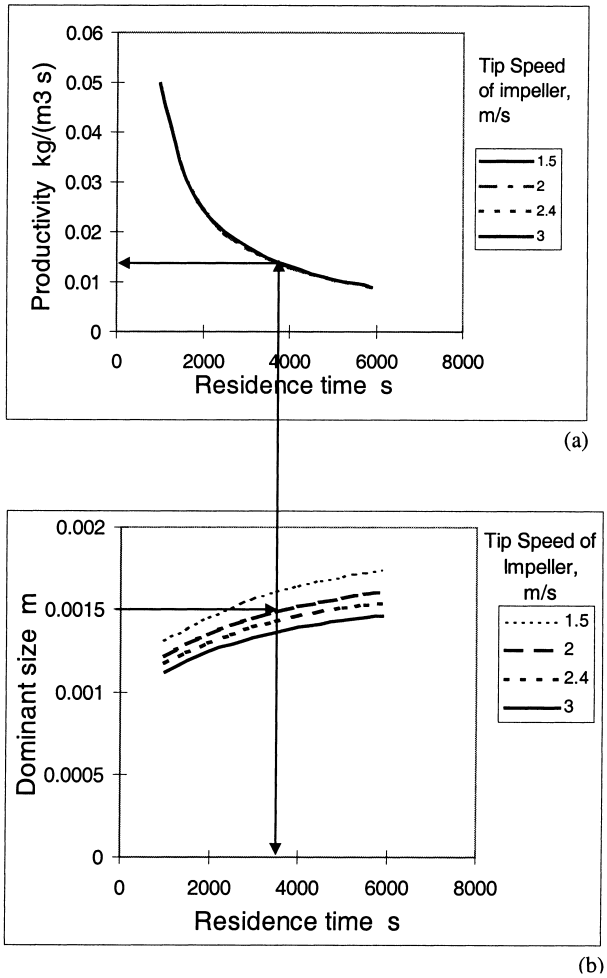

Fig. 8. The design procedure using the neural network: (a) the relationship between residence time and productivity: the tip speeds of the impeller are 1.5–3.0 m/s; (b) the relationship between residence time and dominant size.

tallizer depends on the accuracy of the measured modified Rosin–Rammler number, m, and the parameters in the nucleation rate model. If the modified Rosin–Rammler number, m, was measured based on the particle size distribution produced from a large-scale crystallizer, and the parameters in the nucleation rate model were obtained based on a different scale of a geometrically similar crystallizer, the calculated volume of the crystallizer may be assumed reliable.

8. Conclusions

A neural network for simulating steady state industrial crystallization was developed in this work. The results obtained from the neural network were verified by theoretical equations and experimental data. This neural network can be used to simulate steady state crystallization processes and to design a crystallizer. The accuracy of the simulated results depends on the parameters of the nucleation rate model and on the measured modified Rosin–Rammler number, m. The neural network covers many crystallization systems and might be applied to several continuous operation modes, including cooling crystallization and evaporation crystallization, with fines destruction and classified product removal, size independent crystal growth and size dependent crystal growth. Theoretically, this neural network could be used for any system under any steady state operation conditions, and for any operation mode, if the ranges of the inputs and outputs are those as used in the training.

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