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PHYSICAL PROPERTIES – FORMED DURING SPRAY DRYING – OF MATERIALS WITH THE PROPERTIES OF AN AGGLOMERATE

Ireneusz Zbiciński, Marzena Kwapińska

Faculty of Process and Environmental Engineering, Technical University of Łódź
ul. Wólczańska 213, 93-005 Łódź
e-mail: zbicinsk@ck-sg.p.lodz.pl

Abstract. Systematic studies on the effect of drying and spraying conditions on the physical properties of agglomerate-like materials, i.e. a detergent and cacao, are presented in this paper. A laser PDA technique was applied in order to measure the initial droplet diameter distribution in the spray. The properties of the powders obtained during concurrent spray drying as a function of the temperature of the air drying and the flow rate, the concentration of the raw material and temperature and the initial mean particle diameter in the spray, were analyzed. As a result, it was shown that drying and atomization conditions had no effect on the apparent density of agglomerate-like materials because during drying, particles of a porous structure are formed. It was also found that the bulk density of powders changed when the apparent density of a material or powder particle morphology was changed. The authors propose neural models that predict selected physical properties of powders on the basis of the initial drying and atomization parameters. The proposed neural models well describe the physical properties of the dried products and an extension of the database to other materials would enable the construction of more general models for this group of materials.

Keywords: spray drying, agglomerate-like materials, powder properties, neural networks

INTRODUCTION

One of the most numerous groups of spray-dried materials are agglomerate-like materials [13]. The mechanism of drying these materials has been described by Walton and Mumford [13,14]. Agglomerate-like particles are formed from a suspension. When water is evaporated from sprayed droplets, the material particles present are combined and form agglomerates of a porous structure which enable further moisture evaporation through pores and capillaries. If the inter-particle spaces are large enough to enable the free transport of liquid from the particles to their surface, the agglomerate particles being formed are full and spherical.

If the liquid transport is limited by the pore size, then the surface of the agglomerate particles cracks or behaves like a pseudo-skin and empty particles are formed.

Depending on the drying and atomization conditions, a powder can be obtained that consists of particles characterized by a different morphology which consequently has different physical properties.

In the literature, there are no systematic studies on the effect of drying conditions on the properties of powder and the results available are often ambiguous. There are no publications on the prediction of the properties of the material based on the operating conditions of spray drying.

The aim of this study was to determine the impact of the characteristic properties of this group of materials on the physical properties of spray-dried products and to find quantitative relations between the initial parameters of drying, atomization and the properties of the powder.

SETTING UP THE EXPERIMENT AND THE MEASURING TECHNIQUES

In the Faculty of Process and Environmental Engineering, a drying system was built to enable the relation between the initial drying parameters and the physical properties of powders to be determined. Setting-up the experiment has been discussed in detail in earlier studies [12]. This consists of a concurrent drying tower, a dry air heating system and a much modified feeding system – specially improved for these investigations – that enables control of the raw material temperature and the product receiving system. Dried powder was collected in cyclones. Optic glass was installed in the drying column windows to enable laser measurements of the initial spray parameters (diameter distribution) to be made.

Two agglomerate-like materials were tested, namely cacao and detergent.

The following physical properties of the powders produced were tested: the distribution of the diameter, the apparent density, bulk density and tapped density, the moisture content and the particle morphology as a function of the initial drying and atomization parameters: the mean diameter of the droplets in the spray, the dry air temperature and the flow rate and the concentration and temperature of the raw material. Analysis of the physical properties was made according to Polish standards.

The distribution of the particle size of the powder was determined by a laser particle size distribution analyzer (FRITSCH, Germany).

Bulk density was determined from the mass (m) of constant material volume (V) and calculated from the equation:

$$\rho_b = \frac{m}{V}.$$

The apparent density, i.e. the material density, not taking account of the pores contained inside the material particles [10] was determined by means of a helium pycnometer (AccuPyc 1330, MICROMETRICS).

Microscopic investigations of the powder provided information on the shape and structure of the particles [9].

To reduce the cost of research, preliminary tests were made and the results were analyzed statistically in order to determine which initial drying parameters had the largest influence on the subsequent properties of the powder. The range of investigations was extended for factors that have a significant effect on powder properties. As a result, tests were made for the following range of initial parameters:

- five initial droplet diameters in the spray corresponding to 5 atomization ratios,
- five air drying temperatures: 150, 175, 200, 225, 250°C,
- four air drying flow rates: 200, 300, 400, 500 $m^3 h^{-1}$,
- two dry matter concentrations in the raw material: 30% and 50% for the detergent and 15% and 25% for the cacao,
- two raw material temperatures 30 and 70°C.

In total, 232 experiments were performed. This does not correspond to the product of the number of initial parameters because research for the extended range of parameters was conducted for one concentration and in some drying conditions the product did not dry up.

Table 1 gives the results of laser measurements of the initial diameters of the droplets in the spray.

Table 1. Initial Sauter diameter of spray for detergent and cacao

Atomization ratio	Initial Sauter diameter of spray droplets, D_{32}, μm			
	Feed concentration 30%		Feed concentration 50%	
	Feed temperature 30°C	Feed temperature 70°C	Feed temperature 70°C	Feed temperature 70°C
	Detergent			
0.5	96.5	–	–	–
1	45.8	38.8	48.0	48.6
1.5	40.8	–	–	–
2	38.5	32.9	38.3	33.8
2.5	32.5	30.0	34.8	34.2
	Cacao			
	Feed concentration 15%		Feed concentration 25%	
0.5	88.1	–	61.9	–
1	42.5	38.4	48.4	38.0
1.5	40.8	–	44.5	–
2	34.9	34.6	38.1	35.8
2.5	31.8	32.8	33.6	33.6

RESULTS AND DISSCUSSION

Effect of the initial diameter of the droplets in the spray

The diameter of the particles of powders formed during spray drying depends on the method of atomization, the properties of the raw materials and the drying conditions [7,8].

The effect of the Sauter diameter of sprayed droplets on the Sauter diameter of powder particles is illustrated in figure 1. It was found that an increase in the initial particle diameter in the spray caused an increase in the mean particle diameter of the powders produced. The experiments showed that the mean diameter of the particles of the powder was smaller than the mean diameter of the particles in the spray; this was due to the shrinkage of the particles during evaporation. In certain drying conditions – and only in the case of the cacao powder – the diameter of the particle was larger than the initial diameter of the droplets in the spray; this was related to secondary particle agglomeration [6]. The solid line – in figure 1 – shows the boundary where the diameter of a droplet is equal to the diameter of a particle. Cacao is a multi-component material; among other things, it contains fat which, during drying, moves from inside the particle to the surface. The temperature at which the product was collected, was so high (> 40°C) that the fat present on the surface of the cacao particle had liquefied causing the particles to stick together and stable, secondary agglomerates with much larger diameters to form.

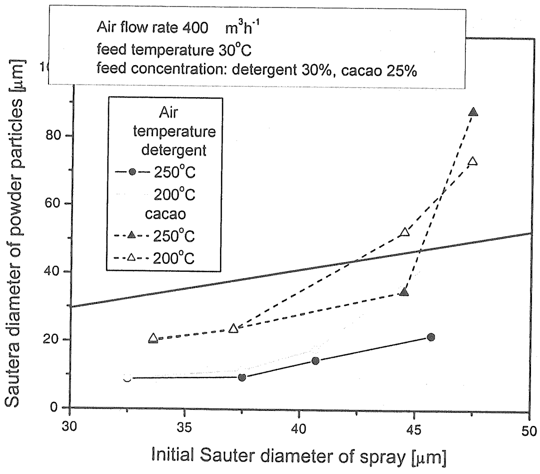

Fig. 1. The effect of the initial Sauter diameter of spray on the diameter of powder particles

Figure 2 shows changes in the bulk density of the materials tested as a function of the initial Sauter diameter of the spray droplets for two air temperatures, 200 and 250°C. It was observed that an increase in the diameter of the droplets in the spray caused an increase in the bulk density of the materials being tested. This effect was caused by inter-particle interactions in the bed of the material and the non-homogeneity of the powder with respect to the diameters of its particle. The initial distribution of the size of the particles in the spray induced distribution both of the diameter in the final product and of the size of the particles – which affects the bulk density of powders [14].

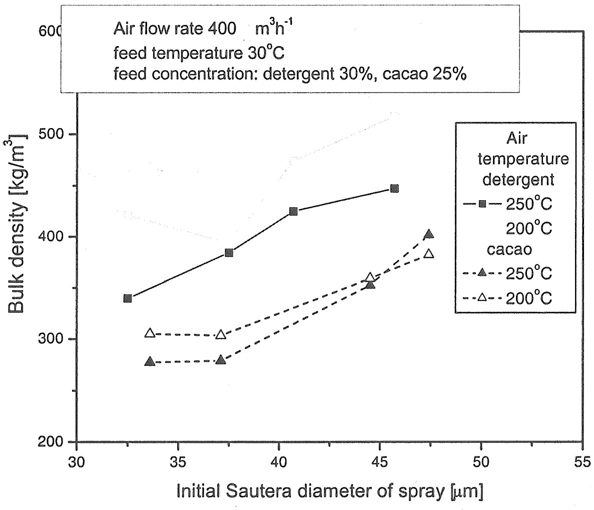

Fig. 2. The effect of the initial Sauter diameter of spray droplets on bulk density

With an increase of particle diameter, inter-particle interactions decrease. Due to the distribution of particle size, the spaces between the larger particles are filled by smaller particles causing better packing of the bed and an increase in the bulk density of the materials. Similar conclusions can be found in the paper by Abdullah and Geldart [1]. Where a significant part of the bed of material – being non-homogeneous in respect of particle diameters – is constituted of particles <20 μm in diameter, the inter-particle forces play a predominant role in the bed packing [3,5].

Particle morphology

A factor that has a significant effect on most physical properties of powders is the morphology of the particles of the material. When analyzing the results of the tests, it was found that the initial parameters of the changes in drying had a small influence on the morphology of the detergent and cacao particles. Both the detergent and cacao produce water suspension; during the evaporation of water from the spray; particles of the material present then stick together and form agglomerates [13].

Figures 3 and 4 show a comparison of the morphologies of the detergent powder tested for the different air flow drying rates and atomization ratios respectively, (the ratio of mass atomizing the air flow rate to the raw material flow rate in the nozzle). Most of the detergent particles were spherical. Fractured particles (fig. 3) occurred only in those powders obtained at a high temperature (225°C, 250°C) and at significant air flow rates (400 $m^3 h^{-1}$, 500 $m^3 h^{-1}$). Characteristically, craters were formed on the surface of the particle of the powder. A relatively small number of destroyed particles was observed. This means that porous particles were formed during drying. This porous structure enabled moisture flow from the inside of the particle to its surface with little diffusion resistance and the absence of mechanical stresses [13].

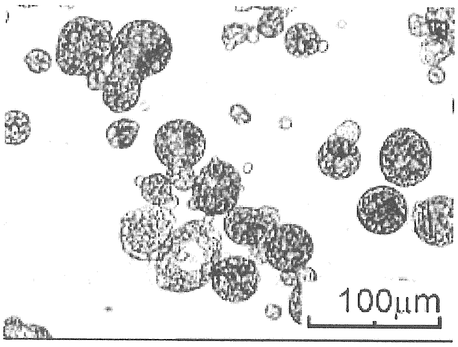

Fig. 3. Morphology of detergent: temperature 250°C, air flow rate 200 $m^3 h^{-1}$, feed concentration 50%, feed temperature 30°C, atomization ratio 1

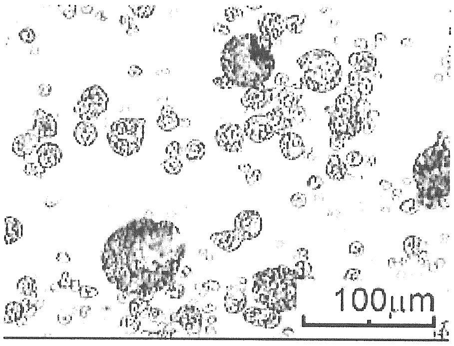

Fig. 4. Morphology of detergent: temperature 250°C, air flow rate 400 $m^3 h^{-1}$, feed concentration 50%, feed temperature 30°C, atomization ratio 2

Figures 5 and 6 illustrate a comparison of the morphology of cacao powder for different drying temperatures and atomization ratios. It was observed that most agglomerate particles were destroyed irrespective of the drying temperature, the air flow rate, the suspension concentration or of the temperature of the raw material. In the conditions in which the drying process was performed, the abrupt evaporation of the moisture from the particles caused their destruction. Moisture transport through the inter-particle pores was insufficient to allow the water present in the particles to evaporate or to cause the water vapor pressure inside the particles to fracture them.

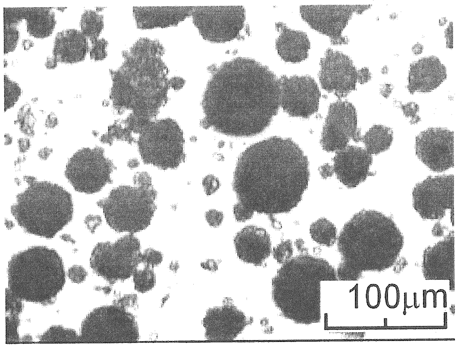

Fig. 5. Morphology of cacao: temperature 200°C, air flow rate 400 m^3h^{-1}, feed concentration 15%, feed temperature 30°C, atomization ratio 1

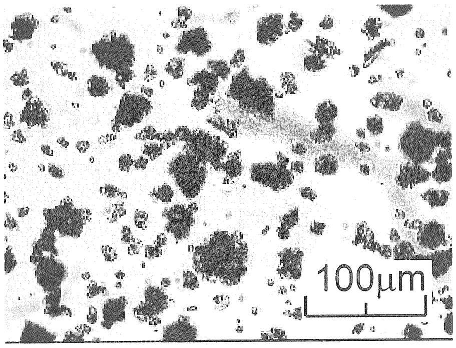

Fig. 6. Morphology of cacao: temperature 250°C, air flow rate 400 m^3h^{-1}, feed concentration 25%, feed temperature 30°C, atomization ratio 2

Drying temperature

The dependence of the apparent density of the materials analyzed on the final temperature of the air drying is shown in figure 7. The drying temperature had no effect on the apparent density of the detergent or the cacao powder. This is due to the nature of these materials and the related mechanism for the removal of moisture. Drying conditions have no influence on the apparent density because during the drying of this group of materials porous particles are formed.

Figure 8 illustrates the dependence of the bulk density of detergent and cacao powder on the air drying temperature for different particle diameters in the spray. The bulk density of these materials depends on the apparent density and packing of the particles in the bed [13]. It was reported that an increase of air temperature from 150 to 250°C caused around a 30% decrease of the bulk density of the detergent. The apparent density of the detergent did not change, while its bulk density decreased; this was related to the morphological changes that occurred during drying and had an influence on the shape of the particle, its surface structure, etc. In powders obtained at the highest drying temperatures, a number of fractured particles were destroyed. The bulk density of such material is smaller because the sharp edges and surface roughness cause the blocking of the particles in the bed, void spaces to form and bulk density to decrease.

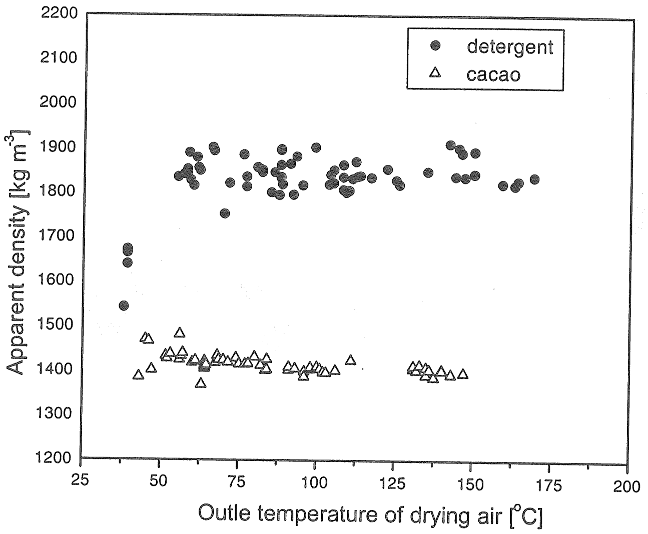

Fig. 7. The effect on the apparent density of the temperature of the air drying at the outlet

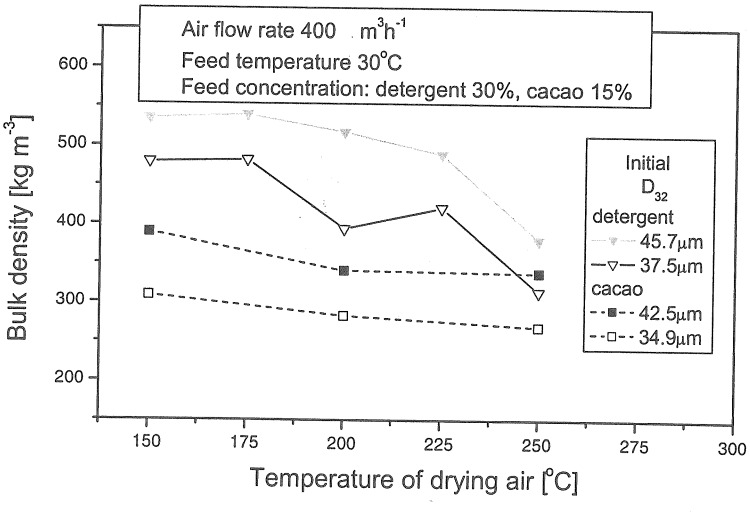

Fig. 8. The effect of the temperature of the air drying on bulk density

The drying temperature has no effect on the bulk density of cacao; the changes observed are within the range of acceptable error. The bulk density of the cacao powder does not change because the apparent density of the material does not change and no changes to particle morphology are observed with changes in the air drying temperature.

Conclusions can be found in the literature that an increase of drying temperature causes a decrease of bulk density in the material [2,4,11], however, it is not always clear which group (skin-forming, agglomerates, crystalline) the material belongs to.

Prediction of the properties of the powder

Neural networks were applied in this study to construct models that would predict the physical properties of powders depending on drying and atomization conditions. Static neural models were developed to predict the basic properties of the powder, i.e. bulk density, tapped density and the Sauter diameter of the particles of the powder. The input vector contained the following initial parameters of drying and atomization: the initial Sauter diameter of particles in the spray, the initial temperature and flow rate of the drying agent, the dry matter content in the raw material and its temperature.

All neural models were built on the basis of the classical *feed-forward* structure with one hidden layer. The transition function was a sigmoid tangent function in the first layer, while in the second layer this was a linear function. The number of neurons in the hidden layer and the number of training epochs were selected by the trial-and-error method. Input and output data were normalized before being introduced to the network. Each network predicted one physical property.

The parameters of the network (the number of neurons in the hidden layer) and the training process (the number of training epochs) in particular neural models, predicting selected material properties are given in table 2. The table also gives the parameters of the statistical estimation of the operation of the network: mean standard error and correlation coefficient.

Figures 9 and 10 show the results of the network simulation for the test data. It was found that the neural models described well the experimental data for bulk and tapped density. Standard errors were relatively small and the correlation coefficients high (tab. 2). An attempt was made to construct common models for both agglomerate-like materials: the cacao powder and the detergent.

The accuracy of the operation of the network was not much worse when compared to the neural models that cover each material separately.

Table 2. Parameters and simulation errors for neural models predicting the properties of detergent and cacao

Powder properties	Number of neurons in the hidden layer	Number of teaching epochs	Arithmetic mean for tested data	Standard error	Determination coefficient R^2
			Detergent		
Bulk density	4	35	454.6 kg m^{-3}	8.1	0.926
Tapped density	6	20	594.7 kg m^{-3}	13.2	0.913
			Cacao		
Bulk density	5	80	377.0 kg m^{-3}	8.89	0.942
Tapped density	5	85	504.5 kg m^{-3}	6.12	0.976

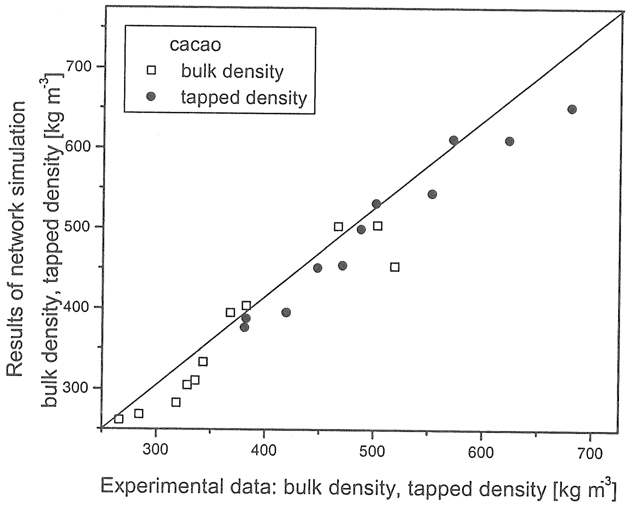

Fig. 9. Prediction of bulk and tapped density of cacao by neural networks

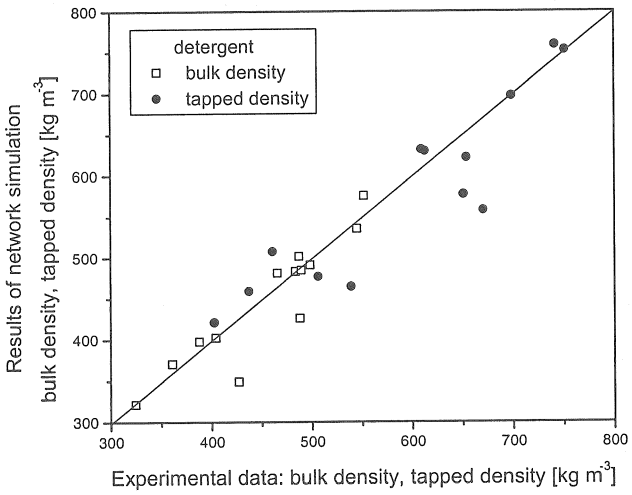

Fig. 10. Prediction of bulk and tapped density of detergent by neural networks

CONCLUSIONS

1. Systematic experimental investigations of the effect of the drying and atomization parameters on the physical properties of agglomerate-like materials during concurrent spray drying showed that the mean diameter of the particles of powder was smaller than the mean diameter of the particles in the spray due to shrinkage of the particles during evaporation. It was observed that an increase in the diameter of droplets in the spray caused an increase in the bulk density of materials induced by inter-particle interactions in the bed of material and the non-homogeneity of the powder with respect to the diameters of particles.
2. The apparent density of agglomerate-like materials (detergent and cacao) does not change in the range of the initial parameters analyzed because during drying, porous particles are formed in the materials.
3. The bulk density of the agglomerate-like materials decreased with the temperature of the drying air. The bulk density of material obtained at the highest drying temperatures was smaller because of the significant changes in particle morphology (sharp edges and surface roughness caused blocking of particles in the bed, the formation of voids and looser packing).

4. Neural models predicting selected powder properties on the basis of drying and atomization conditions were proposed. The proposed neural models describe the selected physical properties of dried products well and an extension of the database to other materials would enable construction of more general models for this group of materials.

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WŁASNOŚCI FIZYCZNE MATERIAŁÓW O STRUKTURZE AGLOMERATU POWSTAJĄCYCH PODCZAS SUSZENIA ROZPRYSKOWEGO

Ireneusz Zbiciński, Marzena Kwapińska

Wydział Inżynierii Procesowej i Ochrony Środowiska, Politechnika Łódzka
ul. Wólczańska 213, 93-005 Łódź
e-mail: zbicinsk@p.lodz.pl

Streszczenie. W pracy przeprowadzono systematyczne badania wpływu warunków operacyjnych procesu suszenia i rozpylania na własności fizyczne materiałów o strukturze aglomeratu: detergentu i kakao. Zastosowano laserową technikę PDA w celu dokonania pomiarów początkowego rozkładu średnic kropel w rozpylanej strudze. Dokonano analizy własności proszków uzyskanych podczas współprądowego suszenia rozpryskowego w funkcji temperatury i natężenia przepływu powietrza suszącego, stężenia i temperatury surówki oraz początkowej średniej średnicy cząstek w strudze. W wyniku przeprowadzonych badań wykazano, że warunki suszenia i atomizacji nie mają wpływu na gęstość objętościową materiałów o strukturze aglomeratu, gdyż podczas suszenia tych materiałów powstają cząstki o porowatej strukturze. Stwierdzono również, że gęstość nasypowa proszków ulega zmianie jeżeli zmienia się gęstość objętościowa materiału albo morfologia cząstek proszku. W pracy zaproponowano modele neuronowe przewidujące wybrane własności fizyczne proszków na podstawie parametrów początkowych suszenia i atomizacji. Modele neuronowe o zaproponowanej architekturze dobrze odwzorowują własności fizyczne produktów suszenia a rozszerzenie bazy danych na inne materiały pozwoliłoby na zbudowanie bardziej ogólnych modeli dla tej grupy materiałów.

Słowa kluczowe: suszenie rozpryskowe, materiały o strukturze aglomeratu, własności proszków, sieci neuronowe