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Modelling of the Spray Coating Process with Biodegradable Polymer Solution for Production of Controlled-Release Fertiliser

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The spray coating process is an important process of the coating particle technology for the production of controlled-release fertilisers (CRFs). Its modelling would be helpful in the production process such as increase material usage efficiency, save production costs, and reduce dispersal waste produced that will cause environmental pollution. This article presents the results of an experimental study to establish an empirical model of the spray coating process with a biodegradable polymer solution. The shape and size of the spray area, droplet size distribution (DSD) and Sauter mean diameter (SMD) were determined by using high-speed camera 1000 frames per second (fps), laser transmitter and the image processing toolbox of MATLAB software. The results showed that the spray area had a cone shape, its mean width, corresponding to a spray distance of 300 mm, was 451 mm at a pressure of 0.18 MPa, viscosity of 50 mPa.s, spray flow of 1.5 mL/min and the droplet size distributions were noted 0.153 to 1.449 mm and droplet SMD was 0.876 mm. The model also showed that droplet SMD was calculated by four dimensionless numbers, will be applied to calculate, optimize and simulate coating particle technology.

1. Introduction

The enhanced efficiency fertilisers (EEFs) is important in modern crop production, which can help to save time, production costs, reduce losses and environmental impact (Timilsena et al., 2015). Controlled-release fertilisers (CRFs) is one of the EEFs and was used widely in agriculture (Trenkel, 2010). CRFs which have a coating layer to control the release of nutrients. They are usually produced using the coating particle technology, that avoids the changing the current production technology (Timilsena et al., 2015), reduces production and investment costs (Trenkel, 2010).

The coating particle technology is applied to form a coating layer which can be classified into 4 types: dry coating, wet coating, melt coating and encapsulation. Among them, the wet coating process is applied the most in industrial production (Saleh and Guigon, 2006). The mechanism of the wet coating process includes spraying and wetting; recrystallization and coated particle (Naz and Sulaiman, 2016). The spray coating process plays an important role in the wet coating process, which decides contact ability of droplets with particles, evaporation of the coating solution and crystallization to form a layer. Determining the mechanism and parameters of the spray coating process will help to adjust the coating process, orient the quality and properties of products.

Many experimental and theoretical models of the spray process have been researched and published. Based on the report of Reitz and Bracco (1986), Lefebvre and Mcdonell (2017) described that drop formation mechanism was classified into four regimes of breakup and shown as Table 1.

Teunou and Poncelet (2002) expanded and described the model of the spray process based on the equation that had been built by Nukiyama and Tanasawa (1939), then Müller and Kleinebudde (2007) also developed that model. It showed that droplet SMD (d_{32}) depends on air velocity, surface tension, liquid density, liquid viscosity and air-to-liquid volume ratio. Varga et al. (2003) presented the mathematical equation for calculating

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the droplet SMD. According to this equation, Aliseda et al. (2008) established and expanded the model. The dimensionless numbers such as Reynolds (Re), Weber (We) and Ohnesorge (Oh) number had been presented in it. Similarly, Yushchenko and Lapin (2011) developed a mathematical equation for the spray process in fire equipment with We, Laplace number (La), the ratio of the viscosity of liquid and gas and dimensionless number characterizes the nozzle design.
Naz et at. (2015) researched to determine the droplet SMD by a high-speed camera and phase Doppler anemometry (PDA) techniques. Recently, Formánek et al. (2019) used a camera system and the image analysis method to determine DSD and SMD in a liquid-liquid dispersions system. This method was also used by Mori (2019) to calculate particle size distributions (PDS).
In this study, by using laser light scattering (LLS) techniques and the image analysis method (IAM), DSD, SMD and the size of the spray area were determined. These results were applied to establish an empirical model of the spray coating process, which was developed on basic models that were previously studied. This model would be more suitable for the coating particle process with biodegradable polymer solution than the other to produce controlled-release urea fertiliser (CRUF).

Table 1: Classification of the breakup regimes

Regime	Description	Predominant drop formation mechanism	Criteria for transition to next regime
1	Rayleigh breakup	Surface tension force	$We_A > 0.4$
2	First wind-induced breakup	Surface tension force; dynamic pressure of ambient air	$We_A > 1.2 + 3.4.Oh^{0,9}$
3	Second wind-induced breakup	Dynamic pressure of ambient air opposed by surface tension force initially	$We_A > 13$
4	Atomization	Unknown	$We_A > 40.3$

2. Materials and methods

2.1 Materials

The materials for synthesis of polymer include: phosphated distarch phosphate (PDSP, E1412) was provided by Nam Bao Tin company, Vietnam; polyvinyl alcohol (PVA, PCT1316, 99 %) was purchased from HiMedia, India; sodium tetraborate ($Na_2B_4O_7.10H_2O$, 99.5 %), glycerol ($C_3H_8O_3$, 99 %) were supplied by Guangdong, China; pure polyacrylic acid emulsion (PAA, 2030) was provided by Nuplex Resins company, Vietnam.

2.2 Synthesis of biodegradable mixing-polymer as coating material solution

Gelatinized 15 g PDSP in 500 mL distilled water at 75 °C, stirring speed of 350 rpm, for 30 min. 0.3 g $Na_2B_4O_7.10H_2O$ was added and kept stirring for 5 min. Then, add slowly 15 g PVA, stir for 30 min, with stirring speed of 450 rpm. The synthesized solution is mixed with polyacrylic acid (PAA) ratio of 3:7 to create a mixing-polymer solution. Next, add 20 mL glycerol and distilled water to adjust the viscosity. The mixing-polymer solution was determined density, viscosity by viscometer Prona RV2 and surface tension by CSC – DuNOUY tensiometer at different temperatures.

2.3 Experimental spray coating process and record images

Diagram of the device to record the images spray coating process was shown in Figure 1a. The whole devices were placed in a closed, dark chamber to avoid light and the impact of environmental factors. Prona RA-100RC-08R nozzle was used for spraying polymer which has a diameter of 0.8 mm, working pressure from 0.1 to 0.3 MPa. Laser transmitter that used to create laser light scattering of droplets, is SDLaser 303 (China), with a wavelength of 532 nm. A digital camera Sony RX100 Mark V (Japan) which has a recording speed of up to , fps, is used to record bright scattering images of droplets during the spray process. The camera and transmitter laser were also exchanged to record images in two dimensions. The experiment was repeated 4 times at an experimental condition and a spray distance (L).
The liquid droplets were created by a pipette with a mixing-polymer solution. Their images, laser scattering images and a standard ruler were recorded with a camera speed of 500 fps to determine a scattering ratio (β).

2.4 Analyze images to determine the size of the droplets and spray area

Videos with a speed of 1,000 fps, which recorded the spray coating process, are transferred to MATLAB software. They were separated into component images to analyze images. Based on bright image contrast, image processing toolbox analyzed and identified pixels with different brightness (Mathworks, 2016). The results of image analysis help to determine the size of the droplets and the spray area.

2.5 Establish an empirical model of the spray coating process by dimension analysis method

The parameters of the spray coating process and their dimensions are presented in Table 2. The dimensionless numbers equation is built based on dimensional analysis methods and Buckingham's pi-theorem (Hanche-Olsen, 2004). It is shown in Eq(1).

Table 2: The parameters of the spray coating process

i	Parameters	Symbol	Unit	Dimension
1	Droplet SMD	d_{32}	m	L
2	Width of spray area	B	m	L
3	Nozzle diameter	d_c	m	L
4	Spray distance	L	m	L
5	Spray velocity	ω_l	m/s	$L.T^{-1}$
6	Density of air	ρ_g	kg/m^3	$M.L^{-3}$
7	Density of solution	ρ_l	kg/m^3	$M.L^{-3}$
8	Viscosity of air	μ_g	Pa.s	$M.L^{-1}.T^{-1}$
9	Viscosity of solution	μ_l	Pa.s	$M.L^{-1}.T^{-1}$
10	Surface tension of solution	σ_l	N/m	$M.T^{-2}$
11	Air flow	Q_g	m^3/s	$L^3.T^{-1}$

$$\frac{d_{32}}{d_c} = A.We^{k_1}.La^{k_2}.\left(\frac{\mu_l}{\mu_g}\right)^{k_3}.\left(\frac{\rho_l}{\rho_g}\right)^{k_4}.\left(\frac{L}{d_c}\right)^{k_5}.\left(\frac{B}{d_c}\right)^{k_6}.\left(\frac{Q_l}{Q_g}\right)^{k_7} \quad (1)$$

Where: A is constant; Weber number, $We = \frac{\omega_g^2.\rho_g.d_c}{\sigma_l}$; Laplace number, $La = \frac{\sigma_l.\rho_l.d_c}{\mu_l}$; spray flow, $Q_l = \frac{\omega_g.\pi.d_c^2}{4}$; the coefficients of equation, k_i (i = 1 – 7).

3. Result and discussion

3.1 Properties of biodegradable mixing-polymer solution

PDSP reacted with PVA, under the action of sodium tetraborate, creating a biodegradable polymer solution. This polymer is mixed with PAA to synthesize a mixing-polymer solution used as a coating material for CRUF (Nguyen et at., 2019). The change of solution parameters according to temperature was shown as Figure 1b.
The surface tension and density of solution change very little with temperature, but its viscosity is too high and change drastically. It must be adjusted with distilled water to achieve a suitable value for the spray process. The results of adjusting the viscosity are described as Table 3

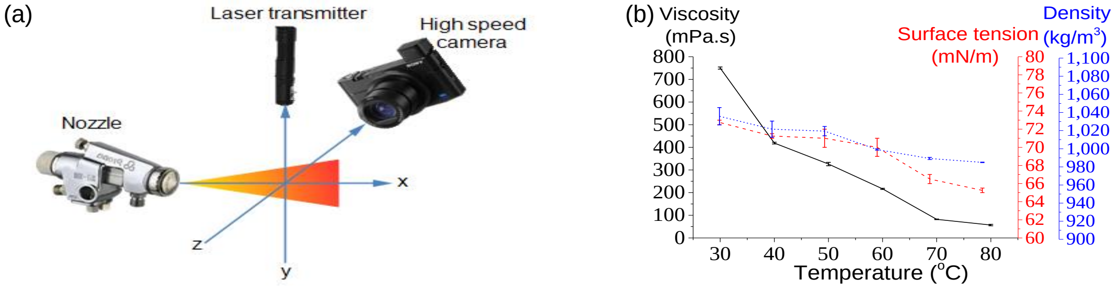

Figure 1: Diagram of the device to record the images (a) and change of polymer solution parameters according to temperature (b)

Table 3: Results of adjusting the viscosity of the solution

No.	Volume PDSP – g – PVA (mL)	Volume PAA (mL)	Volume water (mL)	Viscosity (mPa.s)	Surface tension (mN/m)	Density (kg/m^3)
1	150	350	2.5	115.8	71.0	1,019.4
2	150	350	7.5	63.84	68.5	1,010.3
3	150	350	12.5	50.07	64.6	982.8
4	150	350	15	38.47	62.2	968.3

3.2 Determine the parameters of the spray process by image analysis method

The result on image analysis of the ruler is shown as Figure 2a. The width of the ruler is 67 pixels, corresponding to 25.4 cm (10 inches) in fact, the conversion factor is 0.3791 pixels/mm, which helps to convert the size from pixel to mm unit. Figure 2c and e describe the result on image analysis of droplet and laser light scattering droplet. These results help to determine the scattering ratio, the ratio of the size of the droplet and scattered droplet. The mean scattering ratio with 20 samples is calculated of 1.504, an error is 0.126, standard deviation (SD) of 0.1539, which shows the repeatability and accuracy of the experimental results.

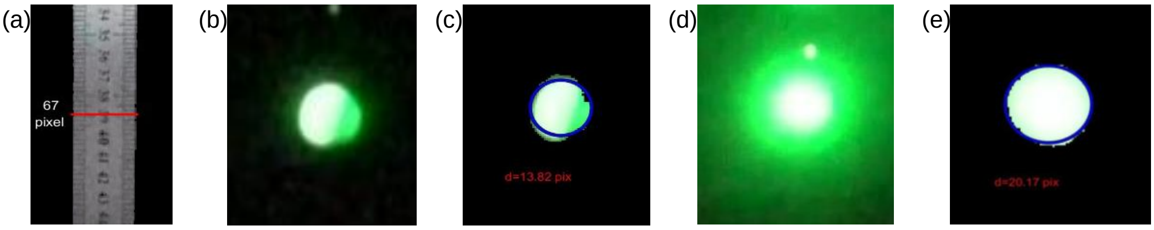

Figure 2: (a) An image of a ruler; (b) an image of a droplet; (c) results on image analysis of droplet; (d) an image of the laser light scattering droplet and; (e) result on image analysis of laser light scattering droplet.

The real-time of a video, which was recorded by the camera at an experimental condition and a spray distance (*L*), was 1 s. This video contains about 1,260 frames and is read by software. About 500 frames were separated and filtered for image analysis. The experimental image of the process and laser light scattering of droplets are described as Figure 3a. The image analysis results of the droplet at the spray distance as 10, 20, 30 cm with Q_l = 2 mL/min, pressure (*P*) of 0.18 MPa, μ_l = 63.84 mPa.s are shown as Figure 3b, c and d.
The nozzle creates a circular spray surface, so the spray area shape is cone with two main size parameters: the spray distance and the width of the spray area. The mean width of the spray area ($\bar{B}$), error and SD at P = 0.18 MPa were calculated and given results in Table 4.

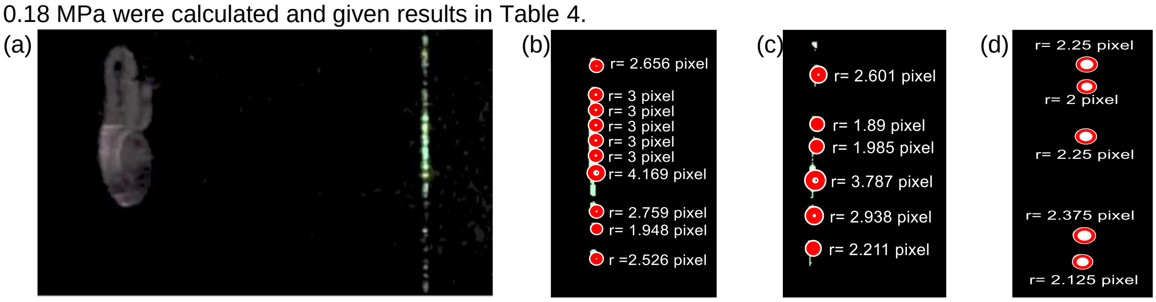

Figure 3: (a) An image of the spray coating process; image analysis results of the droplet at spray distance; (b) L = 10 cm; (c) L = 20 cm (c); (d) L= 30 cm

Table 4: The mean width of the spray area

Exp.	Experimental conditions	L = 10 cm			L = 20 cm			L = 30 cm		
		$\bar{B}, mm$	Error	SD	$\bar{B}, mm$	Error	SD	$\bar{B}, mm$	Error	SD
1	Q = 1.5 mL/min; μ = 63.84 mPas	165.58	15.90	18.656	298.78	24.730	34.227	439.85	15.471	34.660
2	Q = 2 mL/min; μ = 63.84 mPas	206.78	29.334	24.227	273.15	16.356	20.997	361.77	30.426	38.224
3	Q = 1.5 mL/min; μ = 50 mPas	232.43	18.975	22.996	319.25	23.977	29.537	451.15	17.627	20.846

The results in Table 4 show that when the viscosity of solution decreases the size of the spray area is widened because the spray speed increased and the ability to create the droplet is more easily. When the spray flow increases, the spray area is narrowed because the fluid rate is slower while the air flow does not change.
The DSDs are obtained from the image analysis results, corresponding to different experimental conditions and spray distances, described in Figure 4. The SMD decreases as the spray distance increases because the droplets are broken up and dispersed more. As the spray flow increases, the SMD decreases which indicate that the increase of the flow will increase the resistance of fluid, so the SMD and the spray area increase.

Conversely, when the viscosity of solution decreases, the SMD is larger and the spray area is widened, due to the resistance of the fluid decreases in the nozzles.

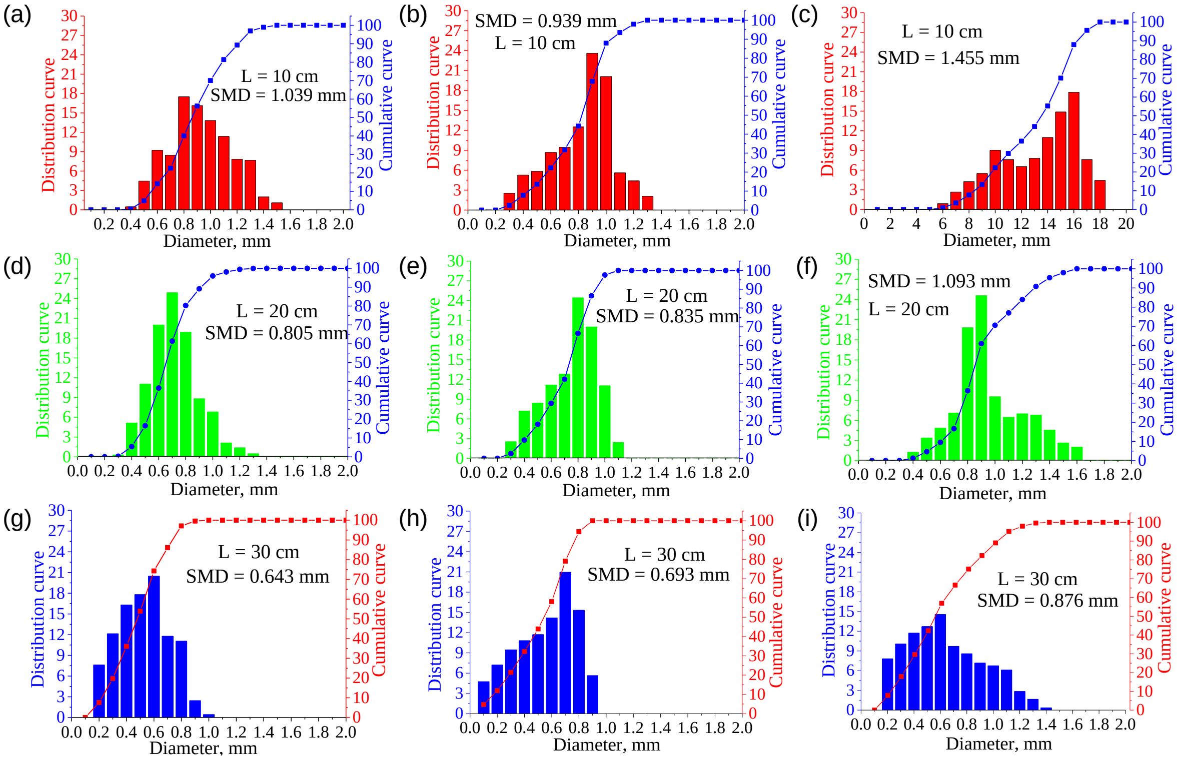

Figure 4: Distribution of droplet size corresponding to spray distances at experimental conditions as Table 4 with experiment No. 1 (a, d, g), No. 2 (b, e, h) and No. 3 (c, f, i)

3.3 Determine the model of the spray coating process

The coefficients of Eq(1) are determined based on the experimental results by using the least-squares regression in Microsoft Excel. The model of the spray coating process is obtained and shown as Eq(2):

$$\frac{d_{32}}{d_c} = 1.62. We^{-0.00565}. La^{0.8714}. \left(\frac{L}{d_c}\right)^{-0.2022}. \left(\frac{B}{d_c}\right)^{-0.2839} \tag{2}$$

The Eq(2) show that the ratio of density, viscosity, flow of liquid and gas do not affect the droplet SMD. Evaluating the calculated from Eq(2) ($(d_{32})_{cal.}$) and the experimental results ($(d_{32})_{exp.}$) are shown in Figure 5.

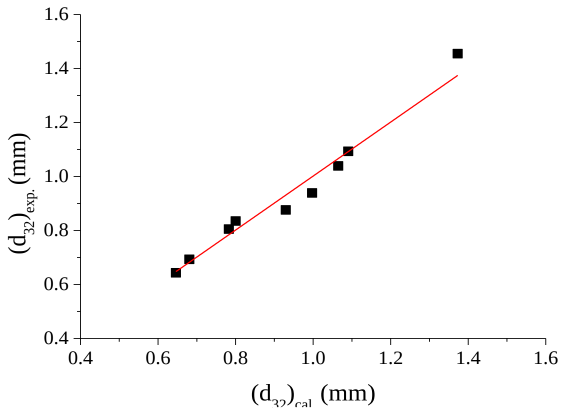

Figure 5: Evaluating the calculated and experimental results

Figure 5 shows that the calculated size of the droplets and experimental SMD are equivalent to each other. The result of evaluating the data compatibility by Fisher distribution also noted the coefficient of determination

R^2 is 0.968, F = 33.6 > $F_{0.95,4,5}$ = 4.757, so they fit. This model can be used to calculate the actual spray coating process and application for similar processes.

4. Conclusions

The results of the study show that the authors selected and determined the method for synthesizing biodegradable mixing-polymer from PVA, PDSP and PAA. This polymer can be used as a coating material to apply in coating particle technology for production of CRUF, which is low cost, environmentally friendly. The parameters of the spray coating process such as the size of the spray area, DSD and SMD were determined by high-speed camera, combining image processing toolbox of MATLAB software. Those results help to 2.5 establish and complete the empirical model of the spray coating process. This is the basis to calculate and control it.

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References

Aliseda A., Hopfinger E.J., Lasheras J.C., Kremer D.M., Berchielli A., Connolly E.K., 2008, Atomization of viscous and non-newtonian liquids by a coaxial, high-speed gas jet. Experiments and droplet size modeling, International Journal of Multiphase Flow, 34(2), 161–175.

Formánek R., Kysela B., Šulc R., 2019, Drop size evolution kinetics in a liquid-liquid dispersions system in a vessel agitated by a rushton turbine, Chemical Engineering Transactions, 74, 1039–1044.

Hanche-Olsen H., 2004, Buckingham's pi-theorem, Mathematical Modelling, NTNU,Trondheim, Norway.

Lefebvre A.H., Mcdonell V.G., 2017, Basic Processes in Atomization, Chapter In: Chigier N. (Ed.), Atomization and Sprays, second edition, CRC Press, Taylor & Francis Group, Boca Raton, USA.

Mathworks, 2016, Image Processing ToolboxTM User 's Guide R2016b, Massachusetts, USA.

Mori Y., 2019, Comparison of particle size distributions of reference particles by using different measuring methods, Chemical Engineering Transactions, 74, 403–408.

Müller R., Kleinebudde P., 2007, Comparison of a laboratory and a production coating spray gun with respect to scale-up, AAPS PharmSciTech, 8(1), 3.

Naz M.Y., Sulaiman S.A., 2016, Slow release coating remedy for nitrogen loss from conventional urea: A review, Journal of Controlled Release, 225, 109–120.

Naz M.Y., Sulaiman S.A., Ariwahjoedi B., 2015, Effect of the borax mass and pre-spray medium temperature on droplet size and velocity vector distributions of intermittently sprayed starchy solutions, The Royal Society of Chemistry, 3704(17), 3704–3714.

Nguyen H.T., Doan V.D., Trinh V.D., 2019, Synthesis of Biodegradable Mixing-Polymer as Coating Material for Controlled-Release Urea Fertilizer, Advanced Materials Research, 1152(3), 43–51.

Nukiyama S., Tanasawa Y., 1939, Experiments on the atomization of liquids in an aircstream, Transactions of the Societyof Mechanical Engineers, 4(14), 86–93.

Reitz R.D., Bracco F.V., 1986, Mechanisms of breakup of round liquid jets, Encyclopedia of Fluid Mechanics, 233–249.

Saleh K., Guigon P., 2006, Coating and encapsulation processes in powder technology, Granulation, 323–375.

Teunou E., Poncelet D., 2002, Batch and continuous fluid bed coating - review and state of the art, Journal of Food Engineering, 53, 325–340.

Timilsena Y.P., Adhikari R., Casey P., Muster T., Gill H., Adhikari B., 2015, Enhanced efficiency fertilisers: A review of formulation and nutrient release patterns, Journal of the Science of Food and Agriculture, 95(6), 1131–1142.

Trenkel M.E., 2010, Slow- and Controlled-Release and Stabilized Fertilizers: An Option for Enhancing Nutrient Use Efficiency in Agriculture, Paris, France.

Varga C.M., Lasheras J.C., Hopfinger E.J., 2003, Initial breakup of a small-diameter liquid jet by a high-speed gas stream, Journal of Fluid Mechanics, 497, 405–434.

Yushchenko P.N., Lapin K.I., 2011, Technical requirements to quality of water dispersion with fire-fighting installations, Mining Rescue, 48, 95–105.