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A B S T R A C T

Liquid atomization is a largely widespread unit operation. The disintegration of a liquid into droplets depends on the nature of the nozzle, on the process parameters as well as physicochemical characteristics of the fluid. The aim of this work is to study the contribution of the process (liquid outlet speed and air pressure) and physicochemical (viscosity and surface tension) factors on the size distribution of droplets generated by single- and two-fluid flat spray nozzles. The obtained droplet median diameters which range between 77 and 594 μm for the single-fluid nozzle and between 11 and 599 μm for the two-fluid nozzle, are discussed in relation with operating conditions of atomization process. Dimensional analysis was performed as a modeling approach. Despite energy input for the droplet formation is known to be influenced by different origins according to single and two-fluid nozzles, it is shown that a unique correlation, with specific values of parameters for each nozzle type, gathers all the parameters affecting droplet size. In the range of process and formulation parameters tested, this correlation is validated and gives satisfactory agreement for the single- and two-fluid nozzles.

Keywords:
Drop
Process control
Multiphase flow
Atomization
Dimensional analysis
Agglomeration

1. Introduction

Liquid spraying is a unit operation largely widespread in many processes and industrial areas: automotive painting, waste incineration unit, pharmaceutical industry (spray-drying, spray congealing, and tablet coating), fuel atomization in combustion processes, mass transfer operations, food processing of granular products in fluid beds, coating of surfaces and particles, *etc.* [1–4]. In the specific case of wet granulation process in mixers or fluid beds, the control of the liquid droplet size distribution is a key step insofar a marked correlation has been found between the droplet size and that of the primary structures (*i.e.* nuclei) that are generated after the impact of the droplet on the powder bed [5]. This step of nuclei formation is of primary importance because it partly controls the extent of wet agglomeration mechanisms during which nuclei are associated in agglomerates [6,7]. Nucleation mechanisms have been described through two parameters [5]: (*i*) the kinetics of droplet penetration inside the powder bed that is largely controlled by formulation properties and (*ii*) the flux of droplets onto the bed surface which corresponds to the physical distribution of liquid in the spray zone and is described by the dimensionless spray flux (ψ_a). The dimensionless spray flux is largely controlled by process parameters, but does not depend on the equipment [5]. The mastering of nucleation mechanisms is related to the values of the dimensionless spray flux, and thus to the fine characterization of sprayed droplet size. This is a challenge in many industries, and especially in couscous production from durum wheat semolina, where the nucleation and agglomeration stage directly controls the efficiency of the production line [8]. Moreover, the influence of the atomization process and formulation parameters on the spray angle is a critical key to manage the agglomeration process, as the spray angle partly determines the spray coverage on the powder bed.

Atomization refers to the disintegration of a liquid into droplets in a surrounding gas. The characteristics of the spray are highly dependent on the spray nozzle type. In single fluid nozzle, the liquid jet is formed thanks to the transformation of the pressure energy into kinetic energy when it crosses the nozzle. In two-fluid nozzles, the pressure of the compressed air is used to disperse the liquid in small droplets thanks to the shear forces which are exerted by the air of atomization on the liquid surface [9]. Two different types of two-fluid nozzles exist considering the position of the mixing nozzles which differs according to whether the compressed air comes into contact with the liquid to pulverize, either in the nozzle (*i.e.* internal mixing) or at the nozzle exit (*i.e.* external mixing) [3]. The phenomenon of atomization generates a high relative speed between the liquid

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Fig. 1. Picture of the single-fluid atomization nozzle (Spraying Systems Co., France).

and the surrounding air. The higher is the relative speed, the higher are the frictional forces, the smaller is the resulting median diameter of the droplet [3]. The first phase of atomization involves the tearing of liquid into filaments and large droplets. The second phase is the breaking of these liquid forms into smaller and smaller droplets [1,10]. Droplet size depends on the nozzle type, process parameters, and physicochemical characteristics of the liquid.

The contribution of physicochemical properties of the liquid has been related to the effects of surface tension, viscosity, and density. It has been demonstrated that an increase in liquid density induces an increase in the droplet size [11]. As the viscosity and the surface tension vary jointly when formulating a liquid, it is still difficult to establish simple correlations between liquid properties and the droplet size. Generally, an increase in viscosity and/or surface tension of the liquid involves a rise in the droplet size [3,9,12–15]. For instance, Rajniak et al. [15] observed an increase in droplet size when increasing the concentration in hydroxypropyl cellulose of the liquid, that could be associated with both increases in viscosity and/or surface tension. It seems necessary to investigate separately the specific effects of the viscosity and the surface tension.

The process parameters (*e.g.* air pressure, liquid flow, spraying angle, temperature, *etc.*) also affect the sprayed droplet size. For instance when using a two-fluid nozzle, it has been shown that an increase in the liquid flow speed, a decrease in the air pressure, and a decrease in the spraying angle induce an increase in the droplet size [3,16]. The variation of the droplet Sauter mean diameter with the air density has been described by a decreasing power law [11]. In the specific case of two-fluid nozzles, the influence of the air pressure overrides that of the liquid flow. The relative pressure of the air (*i.e.* air flow in the nozzle) is the most effective atomization process parameter that affects the droplet size [2].

Many authors have developed empirical or semi-empirical models to predict the droplet size according to the spraying process and liquid physicochemical parameters [17–20]. These models are generally adapted for one nozzle geometry and narrow ranges of process and physicochemical parameters. We did not take note of a model making it possible to predict the size of droplets resulting from both single-fluid and two-fluid nozzles.

The aim of this work is to study the process and physicochemical parameters that control the size distribution of droplets generated by different nozzles. The studied process parameters are the nozzle type (single-fluid or two-fluid nozzles), the liquid outlet speed (from 4.1 to 76.8 $m.s^{-1}$) and the relative air pressure (from 0.5 to 2.5 bar). The selected physicochemical parameters are the fluid viscosity (from 1 to 259 mPa.s) and surface tension (from 42 to 62.5 mN/m). The results are compared and discussed in relation with operating conditions of atomization process. An original adimensional approach is led to mathematically describe the relationships between the droplet median diameter and the process and physicochemical parameters for both the single- and two-fluid nozzles.

2. Materials and methods

2.1. Atomizing systems

A single-fluid and an external two-fluid pneumatic atomizing systems (Spraying Systems Co., France) are used to produce a flat spray for the experiments. The single-fluid system (Fig. 1) can be equipped with two different nozzles (ref. 650017 and 6501), geometrically similar except for the inside diameters of the liquid outlet slit (respectively 0.28 and 0.66 mm). The external two-fluid system is made of a combination of a liquid nozzle and a gas nozzle (Fig. 2). Three external two-fluid systems with three different ratios of the gas outlet area (S_G, which corresponds to the inner area of the central gas outlet minus the outer area of the liquid outlet) to the inner liquid outlet area (S_L) are used: 33.2 (ref. SUJE 416-50 combined with PAJ105-50), 26.3 (ref. SUJE 418-50 with PAJ105-50), or 21 (ref. SUJE 420-50 with PAJ135-50) (Table 1). The higher is the inside diameter of the

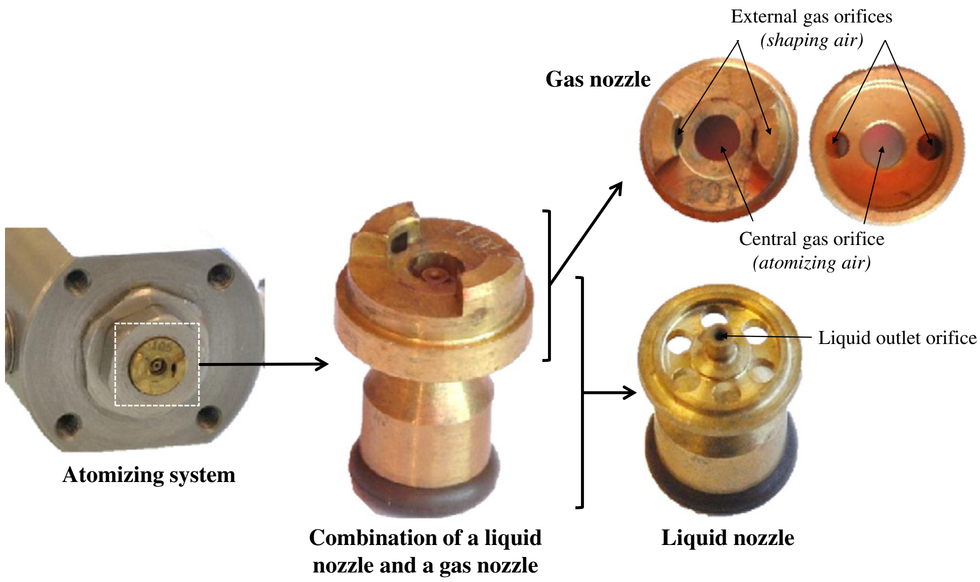

Fig. 2. Pictures of the external two-fluid atomization equipment (Spraying Systems Co., France). The gas outlet area (S_G) corresponds to the inner area of the central gas outlet minus the outer area of the liquid outlet.

Table 1
Principal dimensions of the three two-fluid atomizing systems used in the experiments.

Liquid nozzles	Inside diameters of outlet slits (mm)	Gas nozzles		Gas-to-liquid ratios of the outlet area (S_G/S_L)
		PAJ 105-50	PAJ 135-50	
		2.67	3.43	
SUJE 416-50	0.41	X		33.2
SUJE 418-50	0.46	X		26.3
SUJE 420-50	0.51		X	21

liquid outlet slit, the higher are the liquid flow rates tested during atomization. Although it has been shown that the spray angle and the shape of the spray can be influenced by process parameters (water flow rate, air pressure) and liquid properties [3,9], we consider a constant flat spray angle of 50° (supplier value) for the study.

Atomization experiments were carried out at 20 °C. The liquid is brought to the nozzle at constant flow rate using a peristaltic pump (520S/REM, Watson Marlow, France). Calibration of the liquid flow rate was carried out with water between 15.4 and 26.6 m.s^{-1} for the single-fluid nozzles, and between 4.1 and 76.8 m.s^{-1} under 0.5 bar of relative air pressure for the two-fluid nozzles. It was checked that the liquid flow rate measured at the outlet of the two-fluid nozzles does not change significantly when relative air pressure is increased up to 2.5 bar. The relative air pressure for the two-fluid nozzles is adjusted (between 0.5 and 2.5 bar) using a pressure reducer. The influence of the outlet liquid speed was investigated between 15.4 and 26.6 m.s^{-1} for the single-fluid nozzles and between 4.1 and 76.8 m.s^{-1} for the two-fluid nozzles.

2.2. Pulverized liquids

To study the influence of viscosity and surface tension, experiments were carried out on different ternary mixtures based on deionized water, ethanol, and/or glycerin (Table 2). Ethanol was used to generate different surface tensions because its adsorption kinetic at

Table 2
Composition and physico-chemical properties of the selected atomized liquids (values of density are given ±0.02; values of viscosity are given ±5%).

Nozzle type	Liquid composition (w/w)			Density (kg/m^3)	Surface tension (mN/m)	Viscosity (mPa.s)
	Water	Ethanol	Glycerin			
Single-fluid nozzles	100	0	0	1000	72 (±*1.1*)	1
	96.5	3.5	0	998	62.5 (±*0.1*)	1
	91.3	9.7	0	984	49 (±*0.1*)	1
	81.8	18.2	0	973	41 (±*0.1*)	1
	35.6	4.4	60	1146	49 (±*0.1*)	10.6
	25	0	75	1197	65.3 (±*0.1*)	25.5
	23.3	4.4	72.3	1179	49 (±*0.2*)	25.5
	20.3	10	69.7	1158	39.7 (±*0.2*)	25.5
	15.9	4.4	79.7	–	48.3 (±*0.2*)	51.9
	15.2	0	84.8	1223	63.4 (±*0.1*)	72
	12.8	4.4	82.8	1205	48.4 (±*0.3*)	72.7
	10	10	80	1182	42.5 (±*0.4*)	71.8
Two-fluid nozzles	100	0	0	1000	72 (±*1.1*)	1
	96.5	3.5	0	998	62.5 (±*0.1*)	1
	91.3	9.7	0	984	49 (±*0.1*)	1
	81.8	18.2	0	973	41 (±*0.1*)	1
	15.6	4.4	80	1198	48.6 (±*0.2*)	54.1
	10.5	0	89.5	1235	62.8 (±*0.1*)	134
	4.4	7.9	87.7	1218	49.6 (±*0.2*)	132
	4.5	10	85.5	1194	40.8 (±*0.4*)	132
	4.8	4.4	90.8	1226	49.4 (±*0.3*)	206
	5.9	0	94.1	1247	64.3 (±*0.2*)	259
	3.1	4.4	92.4	1229	50.1 (±*0.2*)	259
	0	10	90	1205	41.6 (±*1*)	259

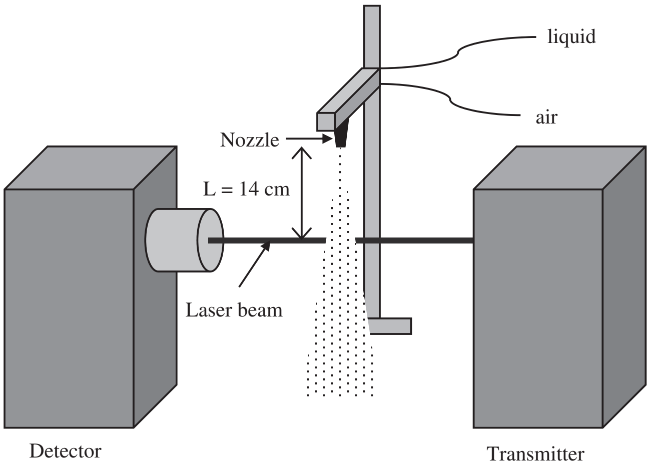

Fig. 3. Sketch of the atomization experiment.

the surface of droplets is instantaneous [10]. Glycerin was selected for its Newtonian behavior and for its aptitude to offer a large range of viscosity in the presence of water and ethanol. The surface tensions of the solutions (between 42 and 62.5 mN/m) were measured in triplicate using a tensiometer Kruss K100 (Kruss, Germany) according to the method of the Wilhelmy plate. The viscosities of the solutions were measured using a rheometer Physica MCR 301 (Anton Paar, Austria) equipped with a double gap mobile. The densities of the solutions were measured using a 50 ml liquid pycnometer. Preliminary tests were conducted to identify the maximal liquid viscosity (80 mPa.s) above which the liquid cannot be atomized using the single-fluid nozzles.

2.3. Droplet size distribution measurement

Liquid droplet sizes were measured by laser diffraction using the Spraytec® (Malvern, France). The extremity of the nozzle is set at a fixed distance L = 14 cm above the laser beam (Fig. 3) as it corresponds to the distance between the nozzle and the powder surface during the wet agglomeration process [7]. Droplet sizes were measured during 2 min and different parameters were calculated. The d_{10}, d_{50}, and d_{90} volume diameters represent the equivalent diameters at which respectively 10, 50 and 90% of the total volume of droplets are inferior. The Sauter mean diameter (d_{32}) is the diameter of a droplet having the same volume-to-surface area ratio as the total volume of all the droplets to the total surface area of all the droplets. The De Brouckere mean diameter corresponds to the mean of the volume distribution (d_{43}). The droplet size span is calculated as $(d_{90}-d_{10})/d_{50}$.

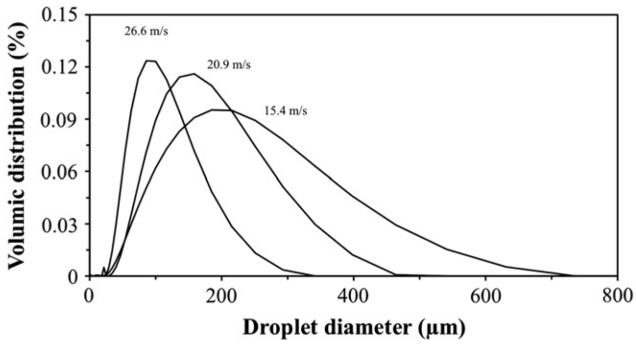

Fig. 4. Droplet size distributions after atomization of pure water using single-fluid nozzles of 0.66 mm inside diameter at outlet liquid speed of 26.6 m/s and 20.9 m/s, and 0.28 mm inside diameter at outlet liquid speed of 15.4 m/s.

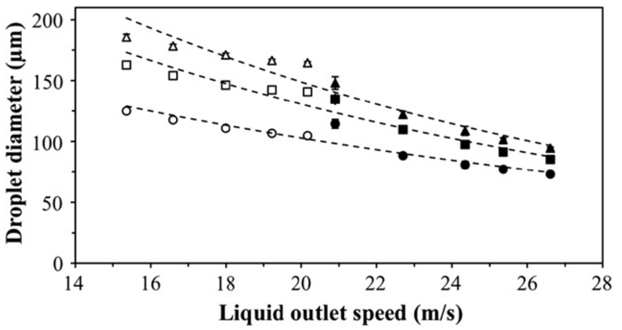

Fig. 5. Influence of liquid outlet speed on droplet diameters (d_{32} (○●), d_{50} (□■), and d_{43} (Δ▲)) after atomization of pure water using two different single-fluid nozzles, at 0.28 mm (white symbols) or at 0.66 mm (blacks symbols) inside diameters.

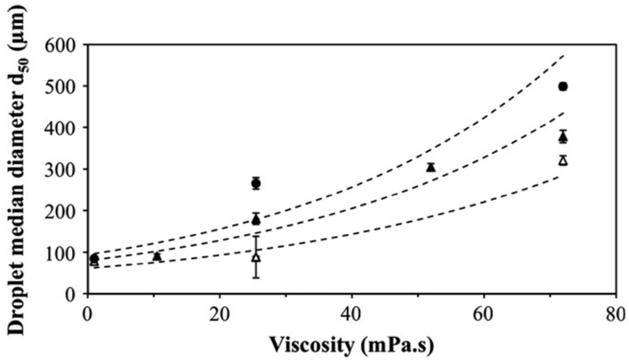

Fig. 7. Influence of liquid viscosity on droplet diameter (d_{50}) after atomization of liquids of different surface tensions (42 mN.m^{-1} (Δ), 49 mN.m^{-1} (▲), and 62.5 mN.m^{-1} (●)) using a single-fluid nozzle (0.66 mm inside diameter) at liquid outlet speed of 26.6 m.s^{-1}.

3. Results and discussions

3.1. Liquid atomization with the single-fluid nozzles

For the single-fluid nozzles, the droplets display monomodal size distributions, whatever the liquid outlet speed (Fig. 4). The measured droplet size distributions were described using different parameters (d_{10}, d_{50}, d_{90}, d_{32}, and d_{43}). As illustrated in Fig. 5, we noticed that the effect of the liquid outlet speed on the droplet diameters (d_{32}, d_{50}, or d_{43}) was almost similar whatever the considered size parameter. As in any case the size distribution was monomodal, we have thus chosen to discuss the influence of physicochemical and process parameters by only displaying the impact on the median diameter (d_{50}) of the sprayed droplets.

3.1.1. Influence of process parameters

When using the single-fluid nozzles, any increase in the liquid outlet speed (from 15.4 to 26.6 m.s^{-1}) induces a large decrease in the droplet diameters (d_{50} varies from 163 to 85 µm) (Fig. 5). In pressure atomization of a liquid using a single-fluid nozzle, pressure is converted to kinetic energy to accelerate the liquid to a high speed, relative to the surrounding ambient air [9,21]. The increase of the liquid outlet speed leads to an increase in both the level of turbulence in the liquid jet and the aerodynamic drag forces exerted by the surrounding air, that both promote the disintegration of the liquid jet in smaller droplets.

3.1.2. Influence of formulation parameters

Under constant outlet liquid flow rate (26.6 m.s^{-1}) and using a low viscosity liquid (1 mPa.s), increases in the fluid surface tension do not affect the droplet diameter (Fig. 6). On the other hand, when using a more viscous liquid (25 or 72 mPa.s), increases in the surface tension induce an almost linear increase of the droplet diameter. For instance for a liquid at 25 mPa.s viscosity, the droplet diameter is tripled when the liquid surface tension increases from 41 to 62.5 mN.m^{-1}.

Whatever the liquid surface tension, we observe increases in the droplet diameter with liquid viscosity (Fig. 7). For instance for a liquid at 62.5 mN.m^{-1} surface tension, the droplet diameter is multiplied by 5 when the liquid viscosity increases from 1 to 72 mPa.s. We thus observed that the ability of the liquid to resist the dynamic forces of atomization is increased with increasing viscosity and/or surface tension. In the case of single-fluid nozzles, an increase in liquid viscosity and/or surface tension has been found to hinder the liquid disintegration and lead to an increase in the amount of energy required for the atomization and also an increase in the droplet diameter [21–24].

3.2. Liquid atomization with the two-fluid nozzles

3.2.1. Influence of process parameters

When using the two-fluid nozzles, measurements of droplet size highlight monomodal droplet diameter distributions (Fig. 8). Increasing the air pressure from 0.5 to 2.5 bar, under constant liquid outlet speed (4.1 m.s^{-1}), induces the formation of smaller droplets and the tightening of the size distribution. When increasing the air pressure, the tightening of the droplet size distribution was observed, whatever the inside diameters of the outlet liquid slit. For instance, when using the two-fluid nozzle with 0.41 mm inside diameter, the diameter span decreases from 2.4 to 1.6 when the relative air pressure varies from 0.5 to 2.5 bar. These effects have been explained by the decrease in the number and volume of large droplets with higher air pressure [16,25].

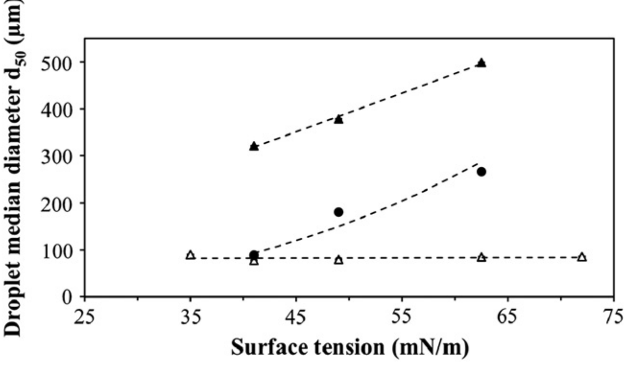

Fig. 6. Influence of liquid surface tension on droplet diameter (d_{50}) after atomization of liquids of different viscosities (1 mPa.s (Δ), 25 mPa.s (●), and 72 mPa.s (▲)) using a single-fluid nozzle (0.66 mm inside diameter) at liquid outlet speed of 26.6 m.s^{-1}.

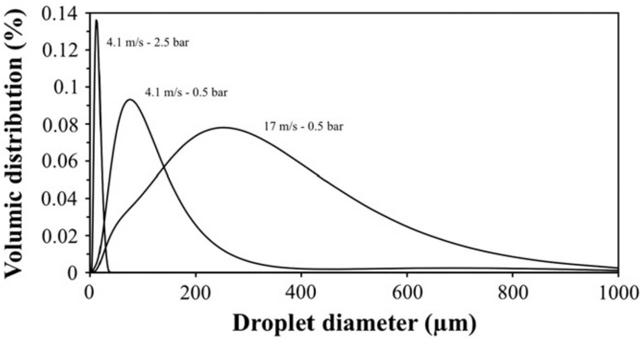

Fig. 8. Droplet size distributions after atomization of pure water using a two-fluid pneumatic nozzle (0.41 mm inside diameter) for two different liquid flow rates and two different atomizing air pressures.

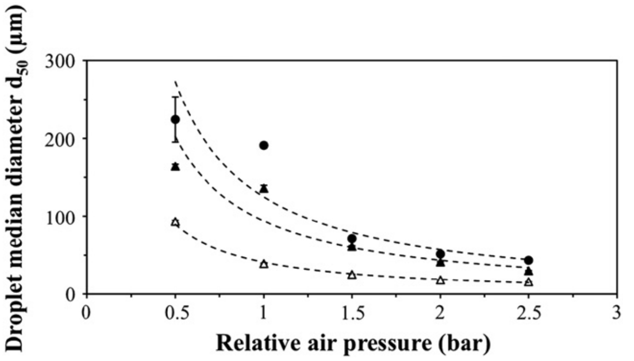

Fig. 9. Influence of relative air pressure on droplet diameter (d_{50}) after atomization of pure water at 10 $m.s^{-1}$ using three different two-fluid nozzles, at 0.41 mm (Δ), at 0.46 mm (▲), or at 0.51 mm (●) inside diameters.

An increase in relative air pressure (from 0.5 to 2.5 bar) also induces a decrease in the droplet diameters (Fig. 9). The relationships between droplet diameters and relative air pressure (P) are described using power law models (Eq. (1)) whatever the liquid outlet speeds.

$$d_{50} \propto P^{-1.11} \tag{1}$$

Increasing the air pressure from 0.5 to 2.5 bar corresponds to an increase in the air outlet speed from 230 to 515 $m.s^{-1}$ which generates an increase in dynamic forces of atomizing air that causes the binder solution to atomize in smaller droplets [3,11,16,23].

On the other hand an increase in liquid outlet speed (from 4.1 to 17 $m.s^{-1}$) under constant relative air pressure (0.5 bar) induces the formation of larger droplets and the broadening of the droplet size distribution (Fig. 8). This effect was observed whatever the inside diameters of the outlet liquid slit (0.41, 0.46, or 0.51 mm) (Fig. 10). For example, the diameter span is tripled when the liquid outlet speed varies from 4.1 to 46 $m.s^{-1}$, at 1.5 bar relative air pressure. Juslin et al. [16] also showed in the case of a pneumatic nozzle, that an increase of the liquid flow rate increases the width of diameter distributions, due to an increase of the amount of large droplets. The dependence of the d_{50} with the liquid outlet speed can be correctly described ($R^2 = 0.947$) using a simple exponential equation (Eq. (2)) whatever the three tested two-fluid nozzles (Fig. 10):

$$d_{50} = exp\left(0.034\, v_{liq}\right) \tag{2}$$

where v_{liq} is the outlet liquid speed ($m.s^{-1}$). The increase in droplet diameters with increasing liquid outlet speed has already been described in the case of two-fluid nozzles [3,23,24].

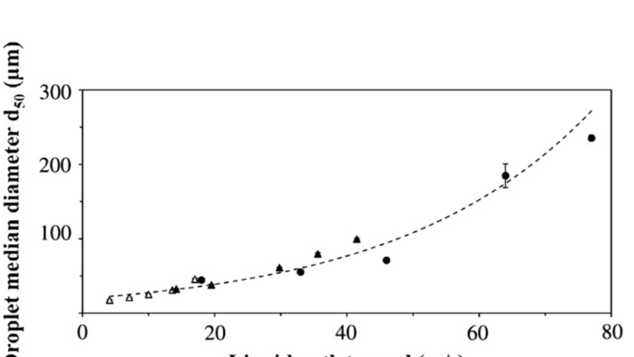

Fig. 10. Influence of liquid outlet speed on droplet diameter (d_{50}) after atomization of pure water using three different two-fluid nozzles, at 0.41 mm (Δ), at 0.46 mm (▲), or at 0.51 mm (●) inside diameters, for an atomizing relative air pressure of 1.5 bar.

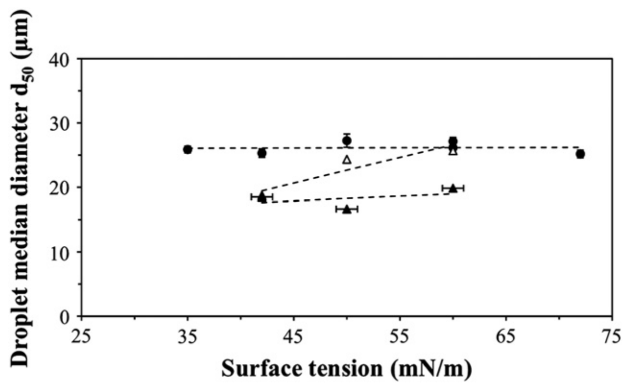

Fig. 11. Influence of liquid surface tension on droplet diameter (d_{50}) after atomization of liquids of different viscosities (1 mPa.s (Δ), 127 mPa.s (▲), and 259 mPa.s (●)) using a two-fluid nozzle (0.41 mm inside diameter) at 10 $m.s^{-1}$ and a relative air pressure of 1.5 bar.

3.2.2. Influence of formulation parameters

Using the two-fluid nozzles, the pulverization of liquid with different physicochemical parameters (viscosity and surface tension) does not seem to affect significantly the droplet size (Figs. 11 and 12). The selected process parameters (liquid outlet speed of 10 $m.s^{-1}$ and relative air pressure of 1.5 bar) seem to overcome the specific influences of the fluid physicochemical parameters and control the fragmentation process.

For the two fluid nozzles, it is still very difficult to establish common rules concerning the influence of viscosity and surface tension on the droplet size. Several authors reported a marked influence of the viscosity on the droplet size. Juslin et al. [16] showed an increase of the droplet mean diameter (from 28.6 to 37.5 µm) with an increase in liquid viscosity (from 1 to 15 mPa.s) using a pneumatic two-fluid nozzle at 1.5 bar relative air pressure and 150 $g.min^{-1}$ liquid flow rate. The ability of a liquid to resist the dynamic force of atomizing air has been supposed to increase with increasing the viscosity, which leads to larger droplet sizes. Rizkalla and Lefebvre [11] and Lefebvre [13] showed an increase in d_{32} with an increase in viscosity for an external mixing prefilming airblast atomizer. Hede et al. [3] also supposed that the effect on d_{32} upon an increase in the viscosity or the surface tension is due to an increase in the amount of energy required to atomize the spray. Other studies showed that the increase in viscosity does not affect the drop size [1,14,26]. Santangelo and Sojka [27] showed that the droplet diameter slightly decreased if the liquid surface tension increased.

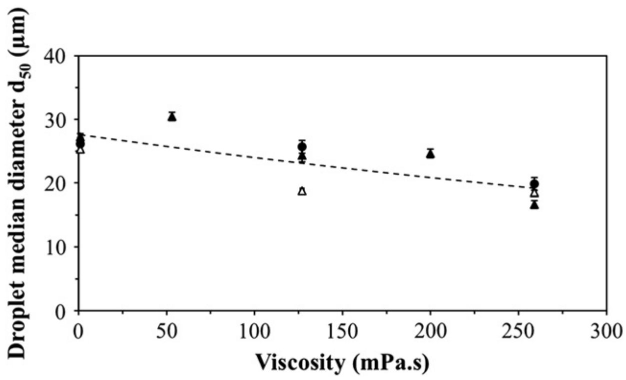

Fig. 12. Influence of liquid viscosity on droplet diameter (d_{50}) after atomization of liquids of different surface tensions (42 $mN.m^{-1}$ (Δ), 49 $mN.m^{-1}$ (▲), and 62.5 $mN.m^{-1}$ (●)) using a two-fluid nozzle (0.41 mm inside diameter) at 10 $m.s^{-1}$ and a relative air pressure of 1.5 bar.

Table 3
Definition of the starting variables and fundamental quantities used for the dimensional analysis of the liquid atomization.

Dimensional variables	Symbol	Fundamental quantities		
		M	L	T
Length between nozzle tip and drop size measurement position	L	0	1	0
Acceleration of gravity	g	0	1	−2
Gas viscosity	μ_G	1	−1	−1
Liquid viscosity	μ_L	1	−1	−1
Gas density	ρ_G	1	−3	0
Liquid density	ρ_L	1	−3	0
Liquid surface tension	σ_L	1	0	−2
Liquid outlet area	S_L	0	2	0
Gas outlet area	S_G	0	2	0
Gas pressure	P_G	1	−1	−2
Liquid outlet speed	V_L	0	1	−1

3.3. Comparison of single-fluid nozzles and two-fluid nozzles

Even if the effect of the liquid outlet speed for two-fluid nozzles (Fig. 10) seems to be opposed to that previously observed for single-fluid nozzles (Fig. 5), the formation of droplets spray is based on the same general mechanism and can be explained by a large differential of velocities between the two phases (air and liquid) whatever the nozzle type.

In the case of two-fluid nozzles, the air speeds (230 to 515 $m.s^{-1}$) are higher than the liquid speeds. An increase in the liquid outlet speed thus induces a decrease in the differential between air and liquid speeds. The lower destabilization energy reported to the liquid volume thus generates droplets of larger size due to the aerodynamic forces (Fig. 10). The relative liquid/air speed is a relevant parameter because it generates superficial waves with short wavelength that cause the fragmentation of the liquid jet [28]. Liquid outlet speed and air pressure thus plays a central role in the fragmentation of the liquid jet. We noticed that under high air pressure conditions, we observe a lower effect of liquid outlet speed on droplet diameter than under low air pressure conditions.

In the case of single-fluid nozzles, these mechanisms remain true. The liquid is sprayed with a high speed in a static ambient air. Any increase in the liquid outlet speed induces an increase in the differential between air and liquid speeds, enhancing frictional conditions and the formation of smaller drops.

In order to explain the specific effect of the viscosity and surface tension on the droplet diameters, it could have been interesting to consider the nature and the geometry of the selected nozzles and achieve a better understanding of the jet rupture mechanisms and drop formation for each nozzle type. Even if a predictive model can be established for a nozzle type under given process and physicochemical conditions, the highlighted correlation could not be directly transposed to other nozzle types and other process conditions. The atomization mechanism, which is specific for each nozzle, makes it difficult to highlight clear tendencies which would express the dependence of the droplet diameters according to the liquid surface tension and viscosity.

3.4. Modeling using dimensional analysis

The originality of our approach lies in the fact that we focus on the droplet diameter resulting from two different types of atomization nozzles (single-fluid and two-fluid nozzles) and different geometries (*i.e.* inside diameters). The tests were carried out on Newtonian fluids to avoid the shearing effect generated by the nozzle. We explored the effect of large ranges of process parameters (*i.e.* liquid outlet speed and air pressure) and physicochemical properties of the liquid (*i.e.* viscosity and surface tension). The air-to-liquid mass flow rate ratios were investigated between 0.06 and 5.11 (range = 5). This range was of the same order of magnitude as the ranges (1.8–15) considered in most of the works dealing with atomization [3,11,17,19,20].

From the experimental study, we aimed in developing an empirical correlation gathering in a unique process relationship all the experimental droplet sizes obtained whatever the operating conditions used (atomization conditions, physicochemical fluid properties, and nozzle geometrical parameters) by using the dimensional analysis [18]. This approach makes it possible to reduce the number of variables describing a physical problem using a set of dimensionless ratios. The number of ratios originally used to describe the problem can be obtained from the Buckingham theorem [29]. The first step of dimensional analysis consists in establishing the list of relevant dimensional parameters. In the case of liquid spraying, the relevant dimensional parameters and their fundamental quantities are listed in Table 3. The Buckingham theorem states that any physically meaningful equation involving n variables expressible in terms of k independent fundamental quantities can be rearranged into an equivalent equation involving a set of p (= n − k) dimensionless variables which are derived as products of powers of the original variables [9,23,29,30]. In our study, we have listed 11 variables (n) that can be expressed in terms of 3 fundamental quantities (k). Following the procedure described in Langhaar [31], we can obtain the dimensional matrix (not shown) that permits to express the dimensionless median diameter according to ‘p = 8’ dimensionless variables (Eq. (3)).

$$\frac{d_{50}}{L} = f\left(\begin{array}{l}\pi_1 = \dfrac{\rho_L}{L^{-1.5}g^{-0.5}\mu_G}; \pi_2 = \dfrac{\rho_G}{\rho_L}; \pi_3 = \dfrac{\mu_L}{\mu_G}; \pi_4 = \dfrac{\sigma}{L^{0.5}g^{0.5}\mu_G}; \pi_5 = \dfrac{S_G}{S_L}; \pi_6 = \dfrac{S_L}{L^2};\\ \pi_7 = \dfrac{P_G}{L^{-0.5}g^{0.5}\mu_G}; \pi_8 = \dfrac{V_L}{L^{0.5}g^{0.5}}\end{array}\right) \tag{3}$$

For each of the two atomization systems (single-fluid nozzles or two-fluid nozzles) the independent influences of the variation of each adimensional number on the value of d_{50}/L were studied. This analysis made it possible to observe that d_{50}/L values (for L = 14 cm) can be mathematically described by the following product of exponential and powers laws (Eq. (4)):

$$\frac{d_{50}}{L} = K\pi_1^{\alpha_1}\pi_2^{\alpha_2}\pi_4^{\alpha_4}\pi_5^{\alpha_5}\pi_6^{\alpha_6}\pi_7^{\alpha_7}\exp(\beta_3\pi_3)\exp(\beta_8\pi_8) \tag{4}$$

Table 4 summarizes the values taken by Eq. (4) parameters according to the two tested spraying system, single-fluid nozzles or two-fluid nozzles. Fig. 13 depicts the adjustment between the values of the dimensionless diameter described by the model (d_{50}/L predicted) and those experimentally obtained for the single-fluid nozzles and the two-fluid nozzles, whatever the process conditions and physicochemical fluid characteristics. We found that the proposed model (Eq. (4)) describes rather well the evolution of the droplet characteristic dimension whatever the nozzle type. Indeed, almost all experimental measurements are contained in a range corresponding to

Table 4
Parameters' value of Eq. (4) for the dimensional analysis of the liquid atomization, according to the spraying system.

Model coefficients	Variables	Mono-fluid nozzles	Two-fluid nozzles
K	–	0.023	$1.66 \cdot 10^{22}$
α_1	Liquid density (π_1)	0.12	0.028
α_2	Gas density (π_2)	0	0.03
β_3	Liquid viscosity (π_3)	$4.34 \cdot 10^{-4}$	$-4 \cdot 10^{-5}$
α_4	Liquid surface tension (π_4)	1.17	0.019
α_5	Gas outlet area (π_5)	0	0.94
α_6	Liquid outlet area (π_6)	0	0.27
α_7	Gas pressure (π_7)	0	−2.2
β_8	Liquid outlet speed (π_8)	−0.045	0.04

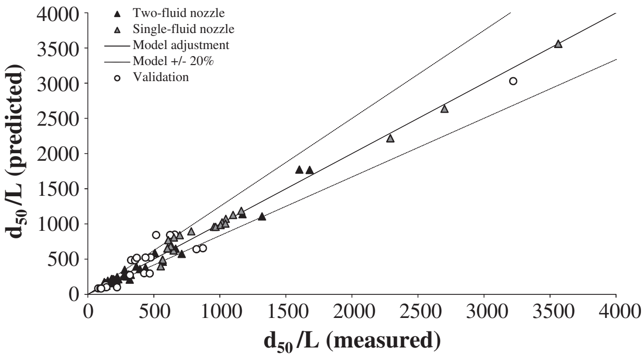

Fig. 13. Predicted *versus* experimental dimensionless droplet diameters for the single- (gray triangles) and the two-fluid (black triangles) nozzles. The droplet size measurement was carried out with a laser beam placed at a fixed distance L = 14 cm from the extremity of the nozzle. Validation points of the model are shown in white circles.

$\pm$20% of the value predicted by the model. The model was validated by tests carried out on the two spraying systems for process and physicochemical properties included in the investigated ranges (Fig. 13).

The values taken by Eq. (4) parameters (Table 4) make it possible to highlight the influence of process conditions. For the two-fluid nozzles, the liquid outlet speed has a direct positive influence on droplet size ($\beta_8 = 0.04$). The air pressure ($\alpha_7 = -2.2$) is the most influencing parameter on the atomization, as the energy allowing the rupture of the liquid jet in drop is brought by the air pressure. The model also takes into account the geometrical aspects associated to the spraying system through the constants α_5 and α_6. Nozzle geometry is shown to have a significant incidence, especially when considering the ratio of the section of the gas outlet to that of the liquid outlet ($\alpha_5 = 0.94$).

On the other hand for single-fluid nozzles, the liquid outlet speed has a negative influence on droplet size ($\beta_8 = -0.045$). The energy allowing the rupture of the jet in droplets only depends on the liquid pressure which is narrowly connected to its outlet speed (Fig. 5). The constants α_5 and α_6 are set to zero because there is no gas outlet, and because the effect of the variation of the nozzle liquid section is taken into consideration through the variation of the liquid outlet speed.

Concerning the influence of the physicochemical properties of the liquid, constants related to the density, surface tension, and viscosity (α_1, β_3 and α_4) are very weak in the case of two-fluid nozzles, reinforcing the fact that process parameters are mainly controlling the droplet size. The influence of density (α_1) is relatively weak. An increase in liquid density generally produces a more compact spray that is less exposed to the atomizing action of the high-speed air and thus lead to a small increase in the droplet diameter [11,13]. On the other hand for single-fluid nozzles, physicochemical parameters highly affect the droplet size. The constant associated with the liquid surface tension ($\alpha_4 = 1.17$) indicates an increase in the energy necessary to spray with an increase in surface tension. The low value of constant α_1 indicates that the influence of density is relatively weak.

The suggested model thus makes it possible to consider the effects of the geometry of the atomizing systems (*i.e.* the liquid outlet slits which are different for the selected nozzles) through a unique equation (Eq. (4)).

4. Conclusions

When using two-fluid nozzles, the atomization mechanisms and the resulting droplet sizes are mainly affected by the air pressure. On the other hand, using single-fluid nozzles, the liquid physicochemical properties and the liquid outlet speed both display significant roles. Although the physical mechanisms subjacent with atomization in the two spraying systems are controlled by the differential speed between the air and the liquid, the different origins of the energy input explain the specific behavior (*e.g.* opposite influence of liquid speed) of the two nozzle types (single-fluid and two-fluid nozzles). According to this mechanism, it has been possible to develop an original and descriptive model based on an adimensional approach. A unique equation was proposed whatever the nozzle type and is associated with two sets of coefficients corresponding to the two types of nozzles. In order to improve the physical understanding of the underlying phenomenon, an experimental approach specially designed to allow the establishment of regime maps through the identification of well-known dimensionless numbers (*e.g.* Reynolds, Weber, *etc.*) could be further carried out.

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