Prediction and Evolution of Drop-Size Distribution of an Ultrasonic Vibrating Microchannel

Moussa TEMBELY [1,*], Christian LECOT[2], Arthur SOUCEMARIANADIN [1]

* Corresponding author: Tel.: (00)33-456-521-120 ; Fax: (00)33-475-561-620; Email: moussa.tembely@ujf-grenoble.fr
1: LEGI-Laboratory of Geophysical and Industrial Fluid Flows-, UMR 5519, University Joseph Fourier, Grenoble, BP 53, 38041 Grenoble cedex
2: Laboratory of Applied Mathematics-, UMR 5127 CNRS, University of Savoie, 73376 Le Bourget-du-Lac Cedex, France

Abstract We report in this paper the evolution of a physically-based drop size-distribution coupling the Maximum Entropy Formalism and the Monte Carlo method to solve the distribution equation of a spray. The atomization is performed by a new Spray On Demand (SOD) device which exploits ultrasonic generation via a Faraday instability. The Modified Hamilton's principle is used to describe the fluid structure/interaction with a vibrating micro-channel conveying fluid excited by a pointwise piezoactuator. We combine to the fluid/structure description a physically based approach for predicting the drop-size distribution within the framework of the Maximum Entropy Formalism (MEF) using conservation laws of energy and mass coupling with the three-parameter generalized Gamma distribution. The prediction and experimental validation of the drop size distribution of a new Spray On Demand print-head is performed. The dynamic model is shown to be sensitive to operating conditions, design parameter and physico-chemical properties of the fluid and its prediction capability is good. We also report on a model allowing the evolution of drop size-distribution. Deriving the discrete and continuous population balance equation, the Mass Flow Algorithm is formulated taking into account interactions between droplets via coalescence. After proposing a kernel for coalescence, we solve the time dependent drop size distribution using a Monte Carlo Method which is shown to be convergent. The drops size distribution upon time shows the effect of spray droplets coalescence.

Keywords: Micro Flow, Spray, Maximum Entropy Formalism, Monte Carlo Method, Coalescence

1. Introduction

Droplet and spray applications are present everywhere in life from industrial operations like fluidized bed to vital natural process as evaporation and sedimentation as well as biotechnology and electronics. Over the past decades, atomization, referring as the disintegration of a bulk liquid material via an atomizer into droplets in a surrounding gas or vacuum with or without a spray chamber, has been extensively developed and applied to a variety of industrial areas such as humidification, medication, pharmaceutical coatings, semiconductor processing, spray drying, and vaporization of volatile anaesthetic agents (Lefebvre, 1989), recent reviews of jet and droplets dynamics and applications can be found in (Eggers and Villermaux, 2008; Yarin, 2006).

Any manufacturing operation that requires the precise metering of materials to specified locations on substrates can take advantage of jet printing. It is well known that piezoelectric jet printing technology is capable of depositing controlled amounts of fluid onto a specified location very accurately. Combining this ability with an increasingly wide selection of fluids has made piezoelectric on-demand jetting system a promising device for developing innovative industrial applications. Jet printing technology offers an amazingly broad range of utilization in wide range areas of applications such as biotechnology, electronics, pharmacology, micro-optics, and many others only limited by our imagination (Calvert, 2001). Four major fluid jetting techniques are well known: Drop On Demand (DOD), Continuous Ink Jet (CIJ), Electro-Valve and Spray Technologies. Our work will focus on an innovative spray technology, which may be qualified as a Spray On Demand (SOD) technology, where spray is generated only if required. This constitutes a major difference with classical spraying technologies where jetting is continuous.

Fluid (ink) jet print-heads offer the advantage of non-contact thus minimizing contamination. Generally classical print-heads are well adapted to applications dealing with Newtonian fluids. However a growing number of engineering applications need robust devices allowing ejection of rheologically complex fluids (like polymer, fluids with particles). In this paper, we give the underlying theory experimental validation for an innovative Spray On Demand printhead (SOD) (Patent,1999).

Some of the numerous applications covered by this device concern the ejection of complex fluids for fuel cell fabrication or metallization (copper and gold electrodes) by electroless plating. The latter application is based on our pending patent and may be used on both impervious and porous substrates.

Despite of its enormous industrial application domain, spray modeling remains a challenge for computational methods and experimental measurements when one wants to predict the drop size distribution. Droplet generation is an extremely complex process that cannot be precisely determined. Current approaches are either semi-empirical or need to be adjusted to each operating conditions. Based on the ultrasonic atomization which is widely used now, we have proposed a physically based drop-size distribution in our previous studies [Tembely et al, 2008]. This approach is necessary for obtaining a specific drop size distribution which can be required in specific applications. In some cases it must have a particular form, narrow or wide, with few small or large drops for some optimizing operation. Small droplet size is desired in spray combustion for rapid heat transfer and vaporization. An ideal atomizer should possess the ability of providing energy-efficient and cost-effective atomization over a wide range of operating conditions. In jet printing, satellite droplets have to be avoided. In the following section we will detail our approach from the SOD fluid/microchanel motion to the evolution of the drop size distribution via Maximum Entropy and Monte Carlo method by using the Mass Flow Algorithm.

2. Vibrating Micro-Channel General Equation of Motion

We consider the system as an approximation of the SOD (Fig.1) shown in Fig. 2. A straight cantilever tube is fixed along the *x*-axis. The fluid enters the tube at the fixed end and exits at the free end. At the middle of the tube is soldered a piezoelectric actuator (PZA) allowing transversal motion of the nozzle. The free end of the tube is beveled. Standard hypothesis for these development assumes, (i) the tube is of uniform annular cross-section, (ii) tube is long compared to its diameter (iii) effect of rotary inertia and shear deformation are ignored, (iv) center line of the tube is inextensible (v) tube is elastic and initially straight, (vi) plane sections remain plane and perpendicular to the beam axis (Bernoulli-Euler beam theory) (vii) effect of internal dissipation and damping are neglected.

It is to be stressed here that our analysis is not for high speed fluid where flutter instability (via a Hopf bifurcation) is observed once the flow velocity exceeds a critical velocity. The dynamical stability analysis and flutter instability control of pipes conveying fluid has been done by many authors (Païdoussis et al.). For our case, let us recall the equation of motion using the elegant Hamilton's Principle taking into account nonlinear effects, print-head configurations, operating conditions, physical and mechanical properties of fluid and structure.

Hamilton formulation seems to be the most convenient for the study it could be formulated as follows [Tembely et al, 2008]:

$$\delta\int_{t_1}^{t_2}\ell dt+\int_{t_1}^{t_2}\delta W dt=\int_{t_1}^{t_2} M_f U \left.\frac{d\vec{r}}{dt}\right|_L \delta\vec{r}_L dt \qquad (1)$$

Where $\vec{r} \approx v(x,t)\vec{j}$ is the location of a material point on the microchannel. And the associated material derivative for fluid /microchannel is defined as $\frac{d\vec{r}}{dt} = \frac{\partial \vec{r}}{\partial t} + U\frac{\partial \vec{r}}{\partial x}$.

ℓ is the Lagrangian of the system, composed of fluid and tube Lagrangian. $v(x,t)$ being the transversal displacement of the microchannel; U the fluid mean velocity, M_f the mass per unit of the fluid.

δW represents the virtual work accounting for forces not included in the Lagrangian due to PZA excitation.

The rhs in (1) represents the momentum transport of the fluid across the open surface of the tube. A simplification of this equation has lead us determining an analytical solution of the tube motion $v(x,t)$ subjected to an external point wise force (Tembely, 2008).

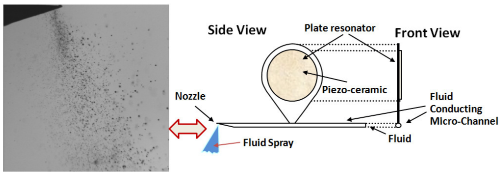

Figure 1: Spray On Demand print-head and visualization of the spray

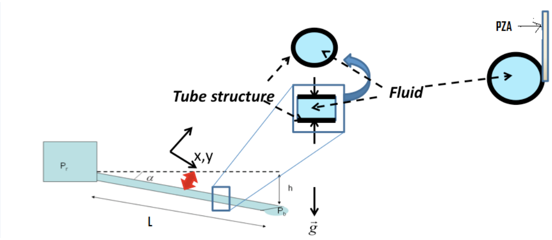

Figure 2: System coordinate and problem statement

3. Prediction of Spray On Demand Drop-Size Distribution

It is very important for spray applications to know the droplet size distribution of the nozzle used. One of the means to describe quantitatively a spray is thus to adopt the tools of statistical analysis. Following (Babinski and Sojka, 2002), there are three methods for modeling drop size distribution: Empirical Method, Probability Function Method and the Maximum Entropy Formalism (MEF) which is the one adopted by us. The aim is to deduce the drop-size distribution by maximizing the Shannon entropy, $S_D = -\sum_{i=1}^{N} p_i \ln(p_i)$, under the constraints detailed hereafter.

3.1 Mass Conservation

Let M_s be the equivalent mass of fluid ejected by the nozzle during a fixed excitation time T_p. We can express M_s differently in the following way:

$$M_s = \rho V_{ol} = \sum m_i = \sum \rho V_i \qquad (2)$$

Where probability p_i is defined as $p_i = N_i / N$. From (2) we deduce the constraint upon mass conservation as

$$\sum_{i=1}^{N} p_i d_i^3 = 1 \quad \text{with} \quad d_i = D_i / D_{30} \qquad (3)$$

For hung drop at the tip of the micro-channel, by applying Newton second law, with a balance between inertial force, surface tension and gravity we deduce:

$$M_s(\bar{\gamma} + g) = \sigma C \cos\theta_E \qquad (4)$$

A good approximation due to Ramanujan for the elliptical nozzle tip circumference is $C \approx \pi[3(a+b) - \sqrt{(3a+b)(a+3b)}]$.

The acceleration and mean acceleration respectively of the nozzle tip could be deduced using (1) as,

$$\gamma(t) = \frac{\partial^2 v(L,t)}{\partial t^2} \quad \text{and} \quad \bar{\gamma}(T_p) = \sqrt{\frac{1}{T_p}\int_0^{T_p} \gamma^2(t)dt} \qquad (5)$$

3.2 Energy Conservation

Instability leading to droplet formation can be viewed as the conversion of the surface, and fluid film kinetic energy, of the hanging drop at nozzle exit, to the droplets surface energy, droplet kinetic energy in addition to the dissipation due to fluid viscosity which is neglected here,

$$E_{surface} + E_{vib} = \zeta_1 E_{droplets} \qquad (6)$$

Expressing $E_{surface} \approx \sigma S_s = \sigma \pi ab$, $E_{vib} \approx \pi M_s f^2 A^2$ and $E_{droplet} = \sum_{i=1}^{N} \sigma S_i = \pi \sigma N \sum_{i=1}^{n} p_i D_i^2$, we deduce the second constraint,

$$\sum_{i=1}^{N} p_i d_i^{\,2} = D_{30} / D_{32} \qquad (7)$$

Using the empirical relationship for wave length (Dobre, 2003) for ultrasonic spray generator, we take the limit amplitude as $A \approx \zeta_2 (\frac{\mu\sigma}{\rho^2 f^3})^{1/5}$ and then deduce from (6, 7), an analytical expression for the Sauter and Volume Mean Diameter, respectively:

$$D_{32} = 1/[\rho \frac{\pi}{6\zeta_1}(\frac{ab(\bar{\gamma}+g)}{\sigma\cos\theta_E C} + \frac{f^2}{\sigma}(\zeta_2 \frac{\mu\sigma}{\rho^2 f^3})^{2/5})] \qquad (8)$$

$$D_{30} = \zeta_3 (\frac{\mu\sigma}{\rho^2 f^3})^{1/5} \qquad (9)$$

Using Lagrange multipliers we deduce the probability by maximizing the Shannon Entropy.

Finally the number-based drop size distribution is deduced using,

$$h_n(D_i) = p_i / \Delta D \qquad (10)$$

where we use the fact diameter space is divided into classes noted as [D_i - $\Delta D/2$, D_i+ $\Delta D/2$].

ζ_i, i=1,2,3, being constants fixed using our model validation by comparing with experiment.

The previous method is limited by the fact that $f_n(D)$ could lead to non physical results (Babinski and Sojka, 2002; Dobre, 2003) where drops of zero sizes seem to exists with a non-zero probability. Whereas the generalized gamma distribution which has been shown to be identical to a Nukiyama-Tanasawa distribution (Dumouchel, 2006), give good results with experiments both for f_n(D) and f_v(D). For our modeling we use the following three-parameter generalized gamma distribution (Dumouchel, 2006):

$$F_n(D) = \frac{q}{\Gamma(\frac{\alpha}{q})} (\frac{\alpha}{q})^{\frac{\alpha}{q}} \frac{D^{\alpha-1}}{D_{q0}{}^{\alpha}} \exp[-\frac{\alpha}{q}(\frac{D}{D_{q0}})^q] \qquad (11)$$

Our previous physically-based model allows computing two of the three parameters of the generalized gamma distribution. The constraint diameter D_{q0} and parameter α are determined using the following relationship,

$$D_{q0} = \left\{ \int_0^{\infty} D^q h_n(D) dD \right\}^{1/q} \qquad (12)$$

And

$$D_{32} = \int_0^{\infty} D^3 F_n(D) dD / \int_0^{\infty} D^2 F_n(D) dD \qquad (13)$$

Where h_n(D) is given using (10). The last parameter q is fixed using experimental validation; this parameter is mainly sensitive to atomization process (Lecompte and Dumouchel, 2008). Thus with this approach, the drop size distribution becomes **dynamic** unlike the classical approach where the parameters have to be adjusted for each operating conditions. The coupling of the previous physical method allow our model to be dynamic and sensitive to physical mechanical and operating conditions of the device.

4. Experimental Validation

We experimentally determine the drop-size distribution of our new print-head using a new shadow imaging technique (Blaisot and Yon, 2005). The only adjustment is made in order to find the different constants of the model. Once these parameters determined, the model can predict the other behavior by changing physical and operating conditions taken into account by the model which has a dynamical capability. It is to be noted that the model takes in account the different physical parameters containing in the mean diameters, micro-channel motion, and also the voltage applied to the PZA and the others parameters required for SOD operation.

The following results are prediction of the model with respect to operating conditions and physical properties of the fluid Fig. 3. As an illustration of the model prediction capability is sketched in Fig. 3b three operating vibrating frequency applied to the PZA. Thus an increase in the working frequency favors the production of smaller drops, where an increase of the working frequency decreases the wavelength and therefore reduces the droplet size. These different results are in good agreement with experimental ones (Dumouchel et al, 2003) for ultrasonic atomizer. In Fig. 3c we have chosen to represent drop-size distribution for 3 different equilibrium contact angle of fluid/structure, such an analysis is out of range for traditional approach of MEF applications. Other capabilities are illustrated in Fig. 3d, where PZA position, a design parameter on nozzle effect is computed.

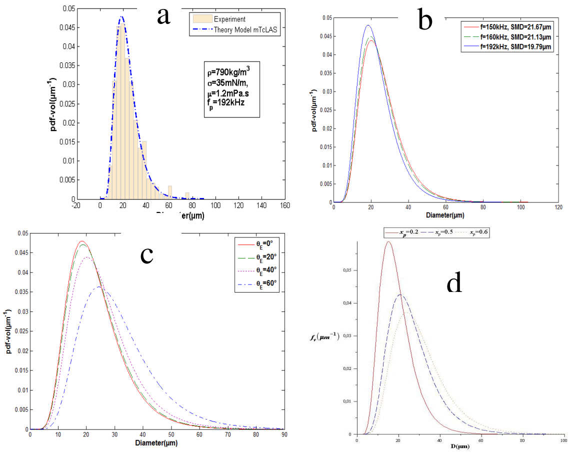

Figure 3: (a) experimental validation; sensitivity to frequency (b) contact angle (c) PZA position effect (d)

5. Continuous Discrete Coalescence Equation

We recall here the drop-size distribution evolution for coalescence (Kocamustafao-gullari and Ishii, 1995)

$$\frac{\partial f_n}{\partial t}(V,t) = \frac{1}{2}\int_{V_{min}}^{V} K_c(V-V',V') f_n(V-V') f_n(V') dV' - f_n(V)\int_{V_{min}}^{V_{max}} K_c(V,V') f_n(V') dV' \quad (14)$$

Here $f(V,\ t)dV$ is the average number of particles of size in $[V,\ V+dV]$ at time t; the function $K(V,\ V')$ is the coalescence kernel describing the rate (or probability) of formation of a particle of size $V + V'$ by coagulation between two particles of size V and V'. The number density $f(V,\ t)$ may

- increase by coalescences of two particles of size $V{-}V'$ and $V' < V$ (first term on the rhs of the equation),
- decrease by coalescence of a particle of size V with any other particle of size V' (second term on the rhs of the equation).

5.1 Mass Flow Formulation

Multiplying equation (14) by V and dividing by,

$$\beta = \int_{V_{\min}}^{V_{\max}} V f dV \quad (15)$$

for normalization purpose, and introducing the mass density function, $m(V,t)=Vf(V,t)$, we obtain, with $g(V,t) = m(V,t)/\beta = Vf/\beta$, the Mass Flow Formulation

$$\frac{\partial g}{\partial t}(V,t) = \int_{V_{min}}^{V} \tilde{K}_c(V-V',V') g(V-V',t) g(V',t) dV' - g(V,t)\int_{V_{min}}^{V_{max}} \tilde{K}_c(V,V') g(V',t) dV' \quad (16)$$

Where $\tilde{K}_c(V,V') = \frac{\beta K_c(V,V')}{V'}$, K_c being the coalescence kernel, given by, see (Tembely et al. 2009)

$$K_c(V,V') = \frac{C_N}{\beta}\pi\frac{(1+\kappa)}{(2+3\kappa)}(D/2+D/2)^2(V^2(D)+V^2(D') - 2V(D)V(D'))^{1/2}\exp[-t_{coal}(V,V')/t_{cont}(V,V')] \quad (17)$$

with viscosity ratio of fluid dand air $\kappa = \mu/\mu_a$. Normalization condition (mass conservation) leads to

$$\int_{V_{min}}^{V_{max}} g(V,t=0) dV = 1 \quad (18)$$

Consequently we have,

$$\int_{V_{min}}^{V_{max}} g(V,t) dV = 1 \quad (19)$$

We apply these results to the physically based drop size distribution of our new Spray On Demand print-head established in section 4. To determine the drop size distribution at initial time, we solve the system for the Maximum Entropy Formalism (MEF), with p_i the discrete probably.

We deduce the physically based distribution (Fig.3) also the initial distribution of our Monte Carlo scheme, using (11) as:

$$g(V,0) = VF_n(V,t=0)/\beta \quad (20)$$

And in discrete form as,

$$g_n(i) = i\Delta V F_n(i\Delta V, t_n)/\beta \quad (21)$$

Initialization

To initiate the computation, we choose N particles

$p_0{}^N(1)$, $p_0{}^N(2)$,…., $p_0{}^N(N) \in N^*$

Such that,

$$\frac{1}{N}\sum_{k=1}^{N}\sigma_{\{i\}}(p_0^{\ N}(k)) \approx g_0(i) \quad (22)$$

Where we denote by $(\sigma_A(i))_{i\geq 1}$ the following sequence for every set $A \subset N^*$:

$$\sigma_A(i) := \begin{cases} 1 \; if \; i \in A \\ 0 \; Othervise \end{cases}$$

Coalescence

We compute number of particles at time t_{n+1} using number particles at time t_n. Let $X_{N,n}^{\ 1}$, $X_{N,n}^{\ 2},\ldots,\ X_{N,n}^{\ N}$ be N independent real random variables uniformly distributed in $\{1,2..N\}$ and $U_{N,n}^{\ k}, 1\leq k\leq N$ be N independent real random variables uniformly distributed on [0,1].

Let us assume that all the random variables $X_{N,n}^{\ k}, 1\leq k\leq N$ and $U_{N,n}^{\ k}, 1\leq k\leq N$ are independent.

The new sizes of particles $P_N^{\ n+1}(k), 1\leq k\leq N$ are defined as:

$$P_N^{\ n+1}(k) = \begin{cases} P_N^{\ n}(k)+P_N^{\ n}(X_{N,n}^{\ k}),\; if\, U_{N,n}^{\ k} < p(P_N^{\ n}(k), P_N^{\ n+1}(X_{N,n}^{\ k})) \\ P_N^{\ n}(k) \quad Otherwise \end{cases} \quad (23)$$

Finally using the Monte Carlo scheme, we solve the evolution of drop-size distribution of our initial distribution submitted to coalescence effect using the mass flow algorithm.

5.1 Monte Carlo Scheme Convergence and Validation

We test the model convergence using $k(i,j)=i+j$, with this kernel an analytical solution for the model exists. As shown in Fig.4, a convergence is obtained for sample of numerical particles of *N*=10000 and time steps of *P*=400.

The initial condition being

$$f_0(i) = \begin{cases} 1 \quad if \quad i=1 \\ 0 \end{cases} \quad (24)$$

And the analytical expression was solving in (Golovin, 1963), and the second moment is given by:

$$M_2(t) = \sum_{i=1}^{\infty} i^2 f(i,t) = e^{2t} \quad (25)$$

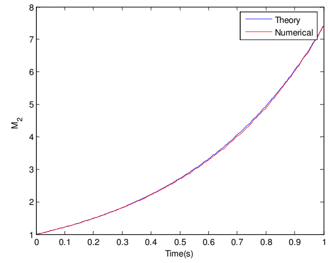

Figure 4: Comparison between analytical and numerical solution of second moment

5.2 Spray Evolution Modeling

We carry out some tests on the evolution of the drop size distribution. We observe that upon time, there is apparition of bigger drop in the spray as shown in Fig.5. The first effect of coalescence is observed at time 50ms. At longer time, we observe the distribution of coalescence effect, with apparition of drops of different size.

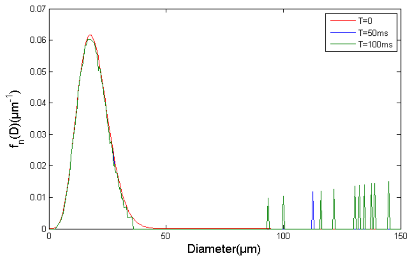

Figure 5: Number based drop size distribution coalescence effect apparition at time 50ms

The following result highlights the volume based-drop size distribution. We see in Fig.6 and Fig. 7, even if there is great number of small droplets, the few big drops represents the majority of the mass of the spray when coalescence occurred which is to be avoided for jet printing application for printing quality.

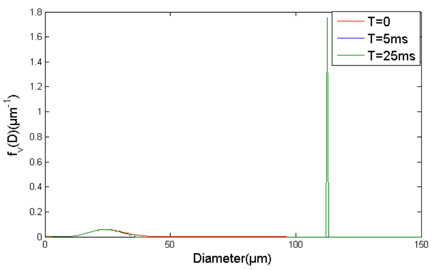

Figure 6: Volume based drop size distribution

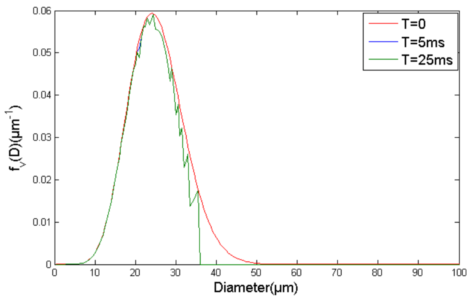

Figure 7: volume based drop size distribution a zoom for small droplets

An improvement of our scheme could be to adopt the Quasi Monte Carlo method (QMC) (Lécot and Wagner, 2004; Lécot and Tarhini, 2007).

Conclusion

In this paper, we have performed a theoretical formulation of instantaneous drop size distribution and its temporal evolution of a new SOD device. Some results have been validated through experiments. Coupling the results from the tube motion conveying fluid established thanks to modified Hamilton's principle, a physically based atomization model has been developed for predicting spray droplet size distribution. Maximum Entropy Formalism (MEF) is then used to predict Drop size distribution using experimental measurements for validation purpose. We have established a physically based prediction model coupling the three parameter generalized gamma distribution and conservation laws of MEF. The model parameters have been deduced using a limited set of experimental measurements obtained with the SOD. This new dynamic model is capable of predicting drop size distribution for well specified operating conditions, fluid and channel structure properties. This new approach avoids the traditional adjustment for each operating condition and has much better predictive capabilities. Indeed, the model being physically-based is capable of predicting the effect of parameters such as equilibrium contact angle between fluid/channel on the drop size distribution, design parameters like PZA position on nozzle and SOD nozzle length. These dynamics sensitivities of the model make our approach a powerful tool for optimizing the promising SOD device. As a final step, the population balance equation, taking into account the interactions between droplets is analyzed. Thus we have established the evolution and resolution of the drop size distribution equation submitted to coalescence effect. In this work we consider only binary interactions where two droplets can coalesce to form a bigger one, this before impacting on the substrate. Based on physical hypothesis, we use our proposed coalescence kernel and couple the model with our previous physically based approach. To solve the problem a Monte Carlo Method which is shown to be convergent is developed highlighting the apparition of new drop due to coalescence. Coalescence preserving the total mass of particles, in order to keep constant the number of numerical particle, we choose The *Mass Flow Algorithm* (MFA) unlike the *Direct Numerical Simulation* (DNS).

As perspective we could improve the method by adopting quasi-Monte Carlo simulation method which consists of replacing the (random) Monte Carlo simulation algorithm by a deterministic. The principle is to replace pseudo-random numbers by deterministic ones.

The device, described in this paper, may be used in a host of innovative microfluidic applications where minute and controlled depositions of highly valued complex fluids are required. Some examples are concern the deposition of metal on substrates or the printing of micro fuel cells.

Nomenclature

a semi-major axis length of nozzle exit shape
b semi-major axis length of nozzle exit shape

g gravity acceleration (=9.81 m.s^{-2})
L length of the nozzle (=5 10^{-2} m)
N total number of drops
N_i number of drops in each class *i*
EI channel (or nozzle) flexural rigidity (=1.8398 10^{-2} SI)
ς inner radius of the tube (=0.5 10^{-3} m)
ρ fluid density
μ fluid viscosity
$\bar{\gamma}$ mean acceleration at the nozzle tip
σ fluid surface tension
θ_E equilibrium contact angle fluid/structure

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