Monte Carlo Method for Physically-Based Drop Size Distribution Evolution

Moussa Tembely, Christian Lécot, Arthur Soucemarianadin

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 coagulation equation. Using the discrete or 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 equation using a Monte Carlo method which is convergent. The evolution of the drop size distribution shows the effect of spray droplets coalescence.

Index Terms— Monte Carlo Method, Maximum Entropy Formalism, Coalescence, Spray, Mass Flow Algorithm.

I. INTRODUCTION

Over the past decades, atomization, defined 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 [1].

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 [2,3]. 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

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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 traditional approaches precise drop size distribution is usually time-independent. Here we propose a modeling of the time-dependent drop size distribution, by using the Maximum Entropy Formalism (MEF). We derive the distribution evolution equation and we use the Mass Flow Algorithm (MFA) for the simulation of the equation with a Monte Carlo method.

II. DROP-SIZE DISTRIBUTION EVOLUTION EQUATION

Due to external environment we assume that the real physical drop size distribution varies with time, expressing for example the effect of coalescence and breakup. The MEF traditionally used ignores the temporal effect on the distribution. In the following section we undertake an analysis combining our physically based MEF approach [2,3] with the balance population method to allow the evolution of the drop size distribution. The different effects to be taken into account for spray droplets are:

- Interactions droplets-gas: evaporation, drag, heat transfer;
- Interactions between droplets: coalescence and breakup.

The evolution of the distribution function is governed by a Boltzmann type equation [4,5]. We focus on the effects of coalescence and breakup and we neglect the evaporation and nucleation phenomena. Using the drop size distribution in a formulation where it depends only on time t, and volume V (instead of the diameter), and assuming a velocity $\vec{V}(V)$ for each droplet of volume V, the balance population equation for the distribution $f_r(V, t)$ can be expressed as :

$$\frac{\partial(Nf_n)}{\partial t} + \nabla . (\vec{V}Nf_n) = R(V) = r_f + r_c \qquad (1)$$

The right-hand side of the equation represents source and sink terms due to fragmentation (r_f) and coalescence (r_c) .

The probability of finding a drop with a volume between V_i and $V_i - \Delta V$ is the same as the probability of finding a drop with a diameter between D_i and $D_i - \Delta D$. The change of formulation between volume and diameter is carried out using the following relationship

$$f_n(D)dD = f_n(V)dV \tag{2}$$

Manuscript received March 22, 2009.

Then we deduce

$$f_n(V) = \frac{2}{\pi D^2} f_n(D) \tag{3}$$

 $f_n(V,t)$ being the number-based drop size distribution to be determined by our analysis. The equation relating number-based and volume-based drop size distribution is given by:

$$f_{\nu}(D) = (\frac{D}{D_{30}})^3 f_n(D)$$
 (4)

The source terms considered are breakup and coalescence. In this work we consider only binary interactions where broken drop splits into two smaller ones and two droplets can coalesce to form a bigger one, this before impacting on the substrate.

General expression for coalescence term could be expressed as:

$$r_{coal}(V) = \frac{1}{2} \int_{V_{min}}^{V} K_c(V - V', V') N(t) f_n(V - V') N(t) f_n(V') dV'$$

$$- N(t) f_n(V) \int_{V_{min}}^{V_{max}} K_c(V, V') N(t) f_n(V') dV'$$
(5)

Let's establish the equation of evolution of the drop size distribution. By adopting a treatment by Discrete Population Balance (DPB) or Classes Method (CM), using a minimum volume V_{min} and a maximum volume V_{max} ; and neglecting advection effect, it means in other word to use a frame moving at spray mean velocity of a given volume containing spray droplets. Defining class q as all the volume between $V_q - \Delta V/2$ and $V_q + \Delta V/2$, from (1), by integrating,

$$n(V_{i},t) = \int_{V_{i}-\Delta V/2}^{V_{i}+\Delta V/2} N(t) f_{n}(V,t) dV \approx N(t) f_{n}(V_{i},t) \Delta V$$
(6)

N(t) being the number total of particles at time *t*. we deduce,

$$\frac{\partial n(V_i, t)}{\partial t} = \Delta V.R(V_i) \tag{7}$$

III. MONTE CARLO METHOD: COALESCENCE EFFECT

Using the previous results the problem can lead to the following equation (8), which could be compared to Smoluchowski equation, the precise resolution of such differential systems of equations is out of range of classical approach. For this reason, we have chosen to use Monte-Carlo methods. We recall the form established,

$$\frac{\partial n_i(t)}{\partial t} = \frac{1}{2} \sum_{k=0}^{l} k_c (i-j,j) n_{i-j}(t) n_j(t) - n_i(t) \sum_{j=0}^{N} k_c (i,j) n_j(t)$$
(8)

Where we set $k_c(i, j) = K_c(V_i, V_j)$.

A. Coalescence Kernel Determination

One the difficulty of our approach is to correctly express, the kernel, here the coalescence of the spray evolution.

From [6] the coalescence kernel could be expressed as a product of the coalescence efficiency and collision frequency,

$$K_{c} = L_{e}(V, V')H_{f}(V, V')$$
(9)

Where we assume the coalescence efficiency could be expressed by

$$L_{e}(V,V') = \exp[-t_{coal}(V,V')/t_{cont}(V,V')]$$
(10)

Where $t_{coal}(V,V')$, $t_{cont}(V,V')$ respectively are average coalescence time of particles of volume V and V', and contact time for the particles. The time required for coalescence could be estimated using

$$t_{coal}(V,V') = C_1 (\frac{R_{V,V'}^{3} \rho_F}{16\sigma})^{1/2}$$
(11)

 C_1 is constant to be adjusted; σ surface tension. $R_{V,V'}$ is the equivalent radius of different radii coalescing and is defined as [7]:

$$R_{V,V'} = \left(\frac{1}{D(V)} + \frac{1}{D(V')}\right)^{-1}$$
(12)

with $V = \frac{\pi}{6}D^3$

The contact time is estimated from [8] for fluid flowing and contribution due to relative velocities between bubbles and assumed here for droplets:

$$t_{cont}(V,V') = \frac{D(V)/2 + D(V')/2}{|u_r(V,V')|}$$
(13)

Where we have neglected turbulent effect. We note $u_r(V,V')$ the relative velocity between drop of volume *V* and *V*', the determination of this relative velocity will be performed as follow.

The relative velocity could be expressed by estimating the limit velocity of falling particles. In fact, we assume in our model that coalescence happens only after this regime is reached, which is quite good since the relaxation time is short.

From Newton second law of falling particles, we obtain

$$R_{ep}^{2}C_{D}(R_{ep}) = \frac{4}{3}Ga$$
 (14)

 R_{ep} , Ga respectively being Reynolds and Galileo (or Archimed) number

$$R_{ep} = \frac{\rho_F V_l D}{\mu_F}$$
 and $Ga = \frac{D^3 g(\rho_p - \rho_F) \rho_F}{{\mu_F}^2}$ (15a)

For spherical fluid particle at low Reynolds number, the Stokes analysis leads to Hadamard-Rybczynski drag law in which the shear stress on the surface induces an internal motion. The drag coefficient becomes,

$$C_D = \frac{8}{R_{ep}} \frac{2+3\kappa}{1+\kappa}$$
(15b)

with viscosity ratio $\kappa = \mu_p / \mu_F$.

From (15a), we deduce, the limit velocity V_l and the relative velocity,

$$\left|u_{r}(V,V')\right| = \left\|\vec{V}_{12}\right\|_{12} = \left(\left(\vec{V}_{1} - \vec{V}_{2}\right)^{2}\right) = \left\|\vec{V}_{1}\right\|^{2} + \left\|\vec{V}_{2}\right\|^{2} - 2\left\|\vec{V}_{1}\right\|_{1}\left\|\vec{V}_{2}\right\|$$
(16)

Where we take the average velocity from velocity directions θ to $\pi/2$.

Then we deduce, the coalescence kernel,

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

With following property verified $k_c(i, j) = k_c(j, i) \ge 0$.

B. Model Equation Reformulation

The method of moment and the size-binning method are valid only for some specific initial distributions and to not describe the precise behavior of the drop size distribution. Monte Carlo seems to be the most advantageous and describe with precision the evolution of the drop size distribution. In order to carry out a precise numerical analysis we will reformulate it in term of mass conservation [9,10,11] and then develop a Mass Flow Algorithm(MFA).

Multiplying by the volume V_i , lead us to mass conservation equation, density of the fluid being considered as constant

$$\sum_{i=0}^{N} V_{i}n(i,t) = \sum_{i=0}^{N} M(i,t) = \sum_{i=0}^{N} M(i,t) = 0 = \frac{m_{0}}{\rho_{F}} = V_{0}$$
(18)

We can normalize the relation by dividing by V_0 the total volume, and we obtain:

$$\sum_{i=0}^{N} \frac{M(i,t)}{V_0} = \sum_{i=0}^{N} m(i,t) = 1$$
(19)

Where we set $m(i,t) = M(i,t)/V_0$,

We have of course, due to mass conservation

$$\frac{d}{dt}\sum_{i=0}^{N}m(i,t) = 0$$
(20)

Multiplying by V_i and using the symmetry of the kernel, we obtain the following equation,

$$\frac{dm_{i}(t)}{dt} = \sum_{j=0}^{i} \tilde{k}_{c}(i-j,j)m_{i-j}(t)m_{j}(t) - m_{i}(t)\sum_{j=0}^{N} \tilde{k}_{c}(i,j)m_{j}(t)$$
(21)

Where we denote

 $\widetilde{k}_{c}(i,j) = \frac{V_{0}k_{c}(i,j)}{V_{j}}$. We can show that \widetilde{k}_{c} is bounded, knowing $K \propto x^{\alpha} \exp(x^{\beta})$. Thus we set $K_{c}^{\alpha} = \sup_{i \neq 1} \widetilde{k}_{c}(i,j)$

C. Monte Carlo Scheme

Once the problem well formulated we look for the drop-size distribution evolution using the Monte Carlo scheme. We choose a fixed time step Δt such that $\Delta t K_c^{\infty} < 1$. We set $t_n = nt$ and $m_i(t_n) = m_n(i)$. For the discrete form we use the finite-difference methods using an explicit Euler scheme to approximate temporal derivative. Using a forward difference at time t_n ,

$$\frac{m_{n+1}(i) - m_n(i)}{\Delta t} = \sum_{j=0}^{l} \tilde{k}_c(i-j,j)m_n(i-j)m_n(j) , i > 1$$
(22a)
$$-m_n(i)\sum_{j=1}^{N} \tilde{k}_c(i,j)m_n(j)$$

Thus, we can deduce $m_{n+1}(i)$:

$$m_{n+1}(i) = \Delta t \sum_{j=0}^{i} \tilde{k}_{c}(i-j,j) m_{n}(i-j) m_{n}(j)$$

$$-(1 - \Delta t \sum_{j=0}^{N} \tilde{k}_{c}(i,j) m_{n}(j)) m_{n}(i)$$
(22b)

Using the mass conservation,

$$\sum_{i=0}^{N} m(j,t) = 1$$
(23)

We express (22b) as:

$$m_{n+1}(i) = \Delta t \sum_{k=0}^{j} \tilde{k}_{c}(i-j,j)m_{n}(i-j)m_{n}(j)$$

$$-\sum_{j=0}^{N} (1 - \Delta t \tilde{k}_{c}(i,j))m_{n}(j)m_{n}(i)$$
(24)

We associate to m_n , the probability P_n defined on N^* :

$$P_n = \sum_{i=1}^{N+1} m_n(i)\delta(i)$$
(25)

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

$$\sigma_{A}(i) \coloneqq \begin{cases} 1 \text{ if } i \in A \\ 0 \text{ Otherwise} \end{cases}$$
(26)

For every $A \subset N^*$

$$\sum_{i=1}^{N+1} m_{n+1}(i)\sigma_{A}(i) = \sum_{i=1}^{N+1} \sum_{j=1}^{i} \Delta t \widetilde{k}_{c}(i-j,j)m_{n}(i-j)m_{n}(j)\sigma_{A}(i)$$
$$-\sum_{i=1}^{N+1} \sum_{j=1}^{N} (1-\Delta t \widetilde{k}_{c}(i,j))m_{n}(j)m_{n}(i)\sigma_{A}(i)$$
$$= \sum_{k=1}^{N} \sum_{j=1}^{N} \Delta t \widetilde{k}_{c}(k,j)m_{n}(k)m_{n}(j)\sigma_{A}(k+j)$$
(27)

$$-\sum_{i=1}^{N}\sum_{j=1}^{N}(1-\Delta t\widetilde{k}_{c}(i,j))m_{n}(j)m_{n}(i)\sigma_{A}(i)$$

Then we deduce,

$$\sum_{i=1}^{N+1} m_{n+1}(i)\sigma_A(i) = \sum_{k=1}^{N} \sum_{j=1}^{N} \{p(i,j)\sigma_A(i+j) + (1-p(i,j))\sigma_A(i)\}m_n(i)m_n(j)$$
(28)

Where we denote $p(i, j) := \Delta t \tilde{k}_c(i, j)$.

The Monte Carlo scheme is then the following.

We choose *N* integers, and for all $n \in N$, we approximate the solution at time t_n by the N following particles denoted,

$$p_n^{N}(1), p_n^{N}(2), \dots, p_n^{N}(N) \in N^*$$

Such that,

$$\frac{1}{N}\sum_{k=1}^{N}\sigma_{\{i\}}(p_n^N(k)) \approx m_n(i)$$
⁽²⁹⁾

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,

$$\frac{1}{N} \sum_{k=1}^{N} \sigma_{\{i\}}(p_0^N(k)) \approx m_0(i)$$
(30)

Coagulation

We compute number of particles at time t_{n+1} using number particles at time t_n .Let

 $X_{Nn}^{1}, X_{Nn}^{2}, \dots, X_{Nn}^{N}$

be N independent real random variables uniformly distributed in $\{1,2...N\}$ and $U_{N,n}^{k}, 1 \le k \le N$ be N independent real random variables uniformly distributed on [0,1].

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

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

defined as:

$$P_{N}^{n+1}(k) = \begin{cases} P_{N}^{n}(k) + P_{N}^{n}(X_{N,n}^{k}), & \text{if } U_{N,n}^{k} < p(P_{N}^{n}(k), P_{N}^{n+1}(X_{N,n}^{k})) \\ P_{N}^{n}(k) & \text{Otherwise} \end{cases}$$
(31)

IV. CONTINUOUS-DISCRETE COALESCENCE EQUATION

We recall here the drop-size distribution evolution for coalescence.

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

$$- f_n(V) \int_{V_{min}}^{V_{max}} K_c(V, V') f_n(V') dV'$$
(32)

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).

Multiplying equation by V and dividing by,

$$\beta = \int_{V_{\min}}^{V_{\max}} V f dV$$
(33)

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

$$\frac{\partial g}{\partial t}(V,t) = \int_{V_{min}}^{V} \widetilde{K}_{c}(V-V',V')g(V-V',t)g(V',t)dV'$$

$$-g(V,t)\int_{V_{min}}^{V_{max}} \widetilde{K}_{c}(V,V')g(V',t)dV'$$
(34)

Where $\widetilde{K}_{c}(V,V') = \frac{\beta K_{c}(V,V')}{V'}$, K_{c} being given by (17).

Normalization condition (mass conservation) leads to

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

Consequently we have,
$$\int_{V_{max}}^{V_{max}} g(V, t) dV = 1$$

We apply these results to the physically based drop size distribution of our new Spray On Demand print-head [2,3]. To determine the drop size distribution at initial time, we solve the following system for the Maximum Entropy Formalism (MEF), with p_i the discrete probably [2,3],

$$S_D = -\sum_{i=1}^N p_i \ln(p_i)$$
(35)

$$\sum_{i=1}^{n} p_i = 1 \qquad \text{with } d_i = \frac{D_i}{D_{30}}$$
(36)

$$\sum_{i=1}^{n} p_i d_i^2 = \frac{D_{30}}{D_{32}}$$
(37)

Where we have shown a physical expression for the Sauter Mean Diameter (SMD) and the volume mean diameter respectively [2]:

$$D_{32} = 1/[\rho \frac{\pi}{6\zeta_1} (\frac{a \not (\gamma + g)}{\sigma \cos \theta_E} C + \frac{f^2}{\sigma} (\zeta_2 \frac{\mu \sigma}{\rho^2 f^3})^{2/5})], \quad D_{30} = \zeta_3 (\frac{\mu \sigma}{\rho^2 f^3})^{1/5}$$

By solving the previous system using an algorithm given by Agmon et al. in [12], we deduce the following numberbased drop size distribution:

$$h_n(D_i) = p_i / \Delta D \tag{38}$$

Our previous physically-based model [2,3] allows computing by coupling with (38) two of the three parameters of the generalized gamma distribution (39). The constraint diameter D_{q0} and parameter α are determined using the following relationship,

$$D_{q0} = \left\{ \int_{0}^{\infty} D^{q} h_{n}(D) dD \right\}^{1/q} \text{ and } D_{32} = \int_{0}^{\infty} D^{3} F_{n}(D) dD / \int_{0}^{\infty} D^{2} F_{n}(D) dD$$
$$F_{n}(D) = \frac{q}{\Gamma(\frac{\alpha}{q})} (\frac{\alpha}{q})^{\frac{\alpha}{q}} \frac{D^{\alpha-1}}{D_{q0}^{\alpha}} \exp[-\frac{\alpha}{q} (\frac{D}{D_{q0}})^{q}]$$
(39)

We deduce the physically based distribution also the initial distribution of our Monte Carlo scheme (Fig.1), using:

$$g(V,0) = V f_n(V,t=0) / \beta$$
 (40)

And in discrete form as,

$$g_i = i\Delta V f_n(i\Delta V) / \beta \tag{41}$$

Using (3) and (39) and finally with the Monte Carlo scheme (31), we solve the evolution of drop-size distribution of our initial distribution submitted to coalescence effect using the MFA.



Figure 1: Physical model drop size distribution Validation of a new Spray On Demand print-head from [2]

V. MODELING RESULTS

A. 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.2, a convergence is obtained for sample of numerical particles of *N*=10000 and time steps of *P*=400.

The initial condition being

$$f_0(i) = \begin{cases} 1 & if \quad i = 1 \\ 0 & (42) \end{cases}$$

And the analytical expression was given in [13], and the second moment is given by:

$$M_{2}(t) = \sum_{i=1}^{\infty} i^{2} f(i,t) = e^{2t}$$
(43)



Figure 2: Comparison between analytical and numerical solution of second moment

B. Spray 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.3. 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 3: 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.4 and Fig. 5, 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 4: Volume based drop size distribution



Figure 5: 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) [11].

VI. CONCLUSION

We have established in this paper 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 determine the 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.

VII. APPENDIX

The convergence of the numerical scheme is proven in the following sense, If

$$\forall i > 1 \quad \lim_{N \to \infty} \frac{1}{N} \sum_{k=1}^{N} \sigma_{\{i\}}(p_n^N(k)) = m_n(i)$$

Then,

$$\forall i > 1$$
 $\lim_{N \to \infty} \frac{1}{N} \sum_{k=1}^{N} \sigma_{\{i\}}(p_{n+1}^{N}(k)) = m_{n+1}(i)$

We introduce Bernoulli random variables,

$$B_{N,n}^{k,i}(k) = \begin{cases} 1 \text{ if } p_n^N(k) = i \\ 0 \text{ Otherwise} \end{cases}$$
$$Y_{N,n}^{i} = \frac{1}{N} \sum_{k=1}^N B_{N,n}^{k,i}(k)$$

Proposition

For every $n \in N$ if

$$\forall i \in N^* \quad Y_{N,n}^{i} \xrightarrow{L^2} m_n(i)$$

Then,

$$\forall i \in N^* \quad Y_{N,n+1} \stackrel{i}{\to} m_{n+1}(i)$$

It can be shown from proposition using the following lemma the convergence of the numerical scheme we present.

Lemma

Let $(X_N)_{N \ge 1}$ be a sequence of real random variables in L^2 and $c \in R$. The following conditions are equivalent:

(i)
$$X_N \rightarrow c$$

(ii) $\lim_{N \to \infty} E[X_N] = c$ and $\lim_{N \to \infty} E[X_N^2] = c^2$

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