CFD Letters www.cfdl.issres.net Vol. 5 (3) – September 2013

Modelling the Formation of Clusters of Drops by Means of the Flocculation and Coalescence Phenomena with Smoothed Particle Hydrodynamics

Alejandro Acevedo-Malavé*

Centro Multidisciplinario de Ciencias, Instituto Venezolano de Investigaciones Científicas (IVIC), *Mérida 5101, VENEZUELA*

Received: 11/10/2012 – Revised 04/12/2012 – Accepted 19/07/2013

Abstract

Smoothed Particle Hydrodynamics (SPH) is a Lagrangian mesh-free formalism and has been useful to model continuous fluid. This formalism is employed to resolve the Navier-Stokes equations by replacing the fluid with a set of particles. These particles are interpolation points from which properties of the fluid can be determined. In this study, the SPH method is applied to simulate the hydrodynamics interaction of many drops, showing some settings for the coalescence, fragmentation and flocculation problem of equally sized liquid drops in three-dimensional spaces. Ranges of value for the collision velocity droplets are chosen, giving rise some different results for the collision of clusters of liquid drops. The velocity vector field formed inside the drops during the collision process is shown.

Keywords: Drops; Coalescence; Collisions; Fragmentation; Navier-Stokes

1. Introduction

The mesh-free Lagrangian particle formalism Smoothed Particle Hydrodynamics (SPH), which is widely used to simulate continuous fluids, is employed to simulate on the three-dimensional space the multiple hydrodynamic coalescence collisions of liquid drops. This formalism is employed for obtaining approximate numerical solutions of the equations of fluid dynamics by replacing the fluid with a set of particles. These particles may be interpreted as corresponding to interpolation points from which properties of the fluid can be determined. Specifically in physics each SPH particle can be treated as a system of smaller particles. The similarity between SPH and molecular dynamics make modeling systems with a complex physics like the emulsion stability problem and the hydrodynamics aspects of the interfacial film when two emulsion droplets collide possible. When two drops collide a circular, flat film is formed, and for sufficiently energetic collisions the evolution of the dynamics leads the interface to be broken giving rise to a bigger drop as a result of coalescence. Examples of the process where the

* Corresponding Author: Alejandro Acevedo-Malavé
Email: alaceved@ivic.gob.ve Telephone: +58 (426) 3690613
 PII: S2180-1363(13)5343-X

coalescence of drops is a very important aspect of the problem are: the combustion of fuel sprays, spray coating, emulsification, waste treatment and raindrop formation [1-4].

Roisman [5] performed a theoretical study on the collision of equalsize drops considering the radial expansion of the droplets, the deformation and stretching of the surface. This author concludes that the deformation of drops in the radial direction is governed by the motion of a ring, while the axial deformation of the drops is by the motion of two globules formed at the end of a stretching liquid jet. Yoon et al. [6] carried out a study about the coalescence of two equal-sized deformable drops in an axi-symmetric flow using a boundary-integral method. The thin film dynamics is simulated up to a film thickness of the order of 10-4 times the non-deformed drop radius. These authors study two different regimes for head-on collisions. At lower capillary numbers the interfaces of the film between the drops remain in a circular flat form up to the film rupture. At higher capillary numbers the film becomes dimpled at an early stage of the collision process as well as the rate of the film drainage slows down after the dimple formation. Eggers et al. [7] conducted a study about the dimensions of the bridge that arises when two droplets collide to form a bigger drop. It is concluded that the analyzed problem is asymptotically equivalent to the twodimensional problem. In addition, these authors study analytically and numerically the case of coalescence of drops in the presence of an external viscous fluid, finding a toroidal structure. Decent et al. [8] studied the formation of a liquid bridge during the coalescence of droplets. These authors consider a model in which the pressure singularity is removed at the instant of the impact for the coalescence of two viscous liquid volumes in an inviscid gas or in a vacuum environment. The formation of the liquid bridge is examined for two cases: (a) two infinitely long liquid cylinders, and (b) two coalescing spheres. In both cases the numerical solutions are calculated for the velocity and pressure fields and the removal of the pressure singularity is confirmed. Rekvig and Frenkel [9] report a molecular simulation study of the mechanism by which droplets covered with a surfactant monolayer coalesce. These authors propose a model such that the rate-limiting step in coalescence is the rupture of the surfactant film. For this numerical study one made use of the dissipative particle dynamics method using a coarse-grained description of the oil, water, and surfactant molecules. It is found that the rupture rate is highest when the surfactant has a negative natural curvature, lowest when it has zero natural curvature, and lying in between when it has a positive natural curvature. Azizi and Al Taweel [10] propose a new methodology for solving the discretized population balance equation (PBE) by minimizing the finite domain errors that often arise when discretizing the drop size domain to study drop breakup and coalescence. Use is made of the size distribution sampling approach combined with a moving grid technique. In addition, an enhanced solution stability algorithm was proposed which relies on monitoring the onset of errors in the various birth and death terms encountered in PBE. This allows for corrective action to be undertaken before the errors propagate in an uncontrollable way. This approach was found to improve the stability and robustness of the solution method even under very high shear rate conditions. Xing et al. [11] put forward a lattice Boltzmann method-based-single-phase free surface model to study the interfacial dynamics of coalescence, droplet formation and detachment phenomena related to surface tension and wetting effects. A perturbation similar to the first step in Gunstensen's color model is added to the distribution functions of the interface cells in order to incorporate the surface tension into the single-phase model. Implementations of the model are verified, simulating the processes of droplet coalescence, droplet formation and detachment from ceiling and from nozzles with different shapes and different wall wetting properties.

Acevedo-Malavé and García-Sucre [12-15] applied the SPH method to model for first time the hydrodynamical coalescence collisions of two equal and unequal size liquid drops in three-dimensional space (on a vacuum environment) in head-on and off-center collisions. As a result of the collision between the droplets, the formation of a circular flat film is observed. A range of values for the droplets collision velocity that produces the permanent coalescence of the drops is chosen. In these references [12-15] three possible outcomes for the collision between the droplets are found which correspond to: permanent coalescence, fragmentation, and flocculation of drops. If

the collision velocity is greater than the range of values for the permanent coalescence, a fragmentation phenomenon is obtained, and if the collision velocity is below this, range flocculation of the drops occur.

In this work, the SPH method is applied to simulate in three-dimensional space multiple hydrodynamics coalescence collisions and formation of clusters of liquid drops in a vacuum environment. This formalism is employed in order to obtain approximate numerical solutions of the equations of fluid dynamics. Here, the model is performed in 3D and it is used the cubic B-spline kernel [16]. In this paper water drops are considered, and the equation of state in the hydrodynamical code was a general Mie-Gruneisen form of equation of state.

2. Numerical details

2.1. The Smoothed Particle Hydrodynamics method

Smoothed Particle Hydrodynamics is a method to solve approximately the equations of fluid dynamics, replacing the fluid by a set of particles. The SPH method was simultaneously invented by Lucy [17] and Gingold and Monaghan [18] to solve astrophysical problems.

In the SPH model, the fluid is represented by a discrete set of N particles. The position of the ith particle is denoted by the vector r_i, i=1,…, N. The SPH scheme is based on the idea that a smoothed representation $A_s(r)$ of the continuous function A(r) can be obtained from the convolution integral

$$A_s(\mathbf{r}) = \int A(\mathbf{r}')W(\mathbf{r}-\mathbf{r}', h)d\mathbf{r}'. \quad (1)$$

Here h is the smoothing length, and the smoothing function W satisfies the normalization condition

$$\int W(\mathbf{r}-\mathbf{r}', h)d\mathbf{r}' = 1. \quad (2)$$

The integration is performed over all spaces. In the limit of h tending to zero, the smoothing function W becomes a Dirac delta function, and the smoothed representation $A_s(r)$ tends to A(r).

In the SPH scheme, the properties associated with particle i, are calculated by approximating the integral in eq. (1) by the sum

$$A_i = \sum_j \Delta V_j A_j W(\mathbf{r}_i - \mathbf{r}_j, h) = \sum_j m_j \frac{A_j}{\circ_j} W(\mathbf{r}_i - \mathbf{r}_j, h). \quad (3)$$

Here ΔV_j is the fluid volume associated with particle j, and m_j and ρ_j are the mass and density of the jth particle, respectively. In the above sum, A_j is the value of a physical field A(r) in the particle j, and the sum is performed over all particles. Furthermore, the gradient of A is calculated using the expression

$$\nabla A_i = \sum_j m_j \frac{A_j}{\circ_j} \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h). \quad (4)$$

In the equation (3), ρ_i/m_i can be replaced by the particle number density $n_i=\rho_i/m_i$, so that

$$A_i = \sum_j \frac{A_j}{n_j} W(\mathbf{r}_i - \mathbf{r}_j, h). \quad (5)$$

The particle number density can be calculated using the expression

$$n_i = \sum_j W(\mathbf{r}_i - \mathbf{r}_j, h), \quad (6)$$

and the mass density is given by

$$\rho_i = \sum_j m_j W(\mathbf{r}_i - \mathbf{r}_j, h). \tag{7}$$

Similarly, the gradient can be calculated using the expression

$$\nabla A_i = \sum_j \frac{A_j}{n_j} \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h). \tag{8}$$

The velocity and acceleration fields are given by [12]

$$\frac{d\mathbf{r}_i}{dt} = \mathbf{v}_i,$$

$$\frac{d\mathbf{v}_i}{dt} = \sum_{j=1}^{N} m_j \left(\frac{\sigma_i}{\rho_i^2} + \frac{\sigma_j}{\rho_j^2} \right) \cdot \nabla W_{ij}^h, \tag{9}$$

where σ is the total stress tensor.
The internal energy evolution is given by the expression [12]

$$\frac{dE_i}{dt} = \frac{1}{2} \sum_{j=1}^{N} m_j \left(\frac{p_i}{\rho_i^2} + \frac{p_j}{\rho_j^2} \right) (v_i - v_j) \frac{\partial W_{ij}}{\partial x_i} + \frac{\mu_i}{2\rho_i} \varepsilon_i \varepsilon_i, \tag{10}$$

In the above equation p is the pressure, μ is the dynamic viscosity and ε is the shear strain rate.

In the present work, the model is performed in three dimensions and the cubic B-spline kernel is used [16], which has the functional form:

$$\overline{f}(x,h) = \sum_{-\infty}^{+\infty} [f(x_j)] M_n(x - x_j, h),$$

where M_n is the nth central B-spline.

In this paper, water drops are considered and the equation of state in the hydrodynamical code was a general Mie-Gruneisen form of equation of state with different analytic forms for states of compression (ρ/ρ0-1)>0 and tension (ρ/ρ0-1)<0 [12].

This equation has several parameters, namely the density ρ, the reference density ρ_0, and the constants A_1, A_2, A_3, C_1 and C_2. This equation of state defines the pressure P as

$$P = A_1 \left(\frac{\rho}{\rho_0} - 1 \right) + A_2 \left(\frac{\rho}{\rho_0} - 1 \right)^2 + A_3 \left(\frac{\rho}{\rho_0} - 1 \right)^3 \quad if \quad \left(\frac{\rho}{\rho_0} - 1 \right) > 0 \tag{11}$$

and

$$P = C_1 \left(\frac{\rho}{\rho_0} - 1 \right) + C_2 \left(\frac{\rho}{\rho_0} - 1 \right) \quad if \quad \left(\frac{\rho}{\rho_0} - 1 \right) < 0. \tag{12}$$

In all calculations the following values are used for the constants: A_1=2.20x10^6 kPa, A_2=9.54x10^6 kPa, A_3=1.46x10^7 kPa, C_1=2.20x10^6 kPa, C_2=0.00 kPa, and ρ_0=1000.0 Kg/m^3.

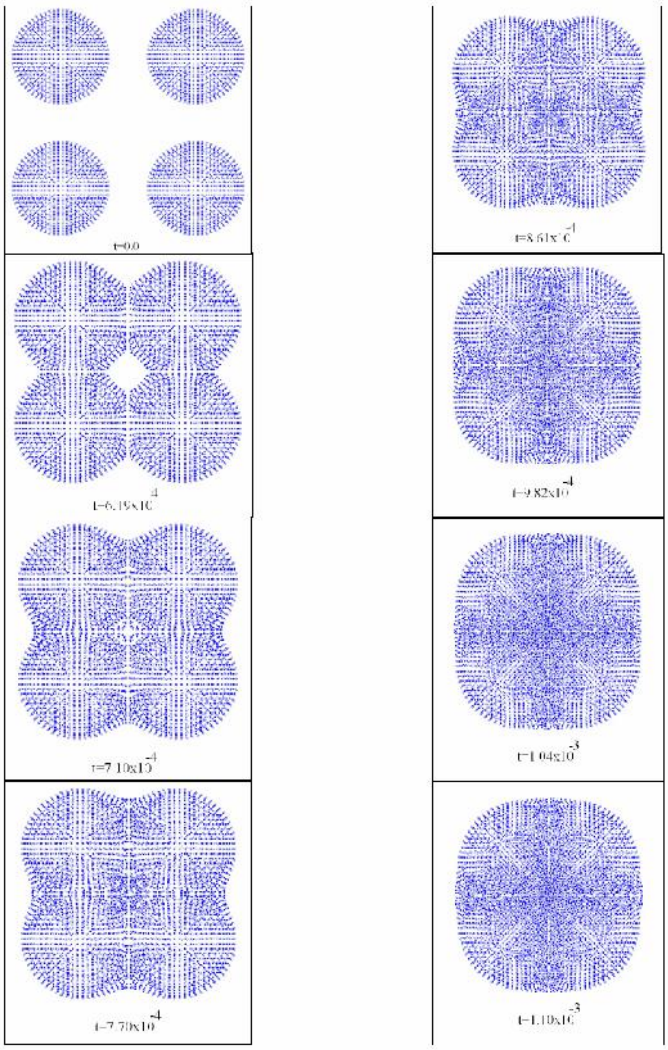

Figure 1. Sequence of times showing the evolution of the collision between four drops (permanent coalescence) with V_{col} = 15.0 mm/ms. The evolution of time is given in milliseconds.

3. Results and discussion

3.1 Coalescence and fragmentation of smaller systems of drops

In order to model the collision of four liquid drops several calculations were carried out, varying the velocity of collision for the modeling of the permanent coalescence of droplets in three dimensional space (3D) using the Smoothed Particle Hydrodynamics method. To do this, within the SPH code were defined drops with diameter of 30μm and 5512 SPH particles for each drop with a collision velocity of 15.0 mm/ms directed to the center of mass of the system.

It can be seen in figure 1 that at t=6.19x10^{-4} ms a flat circular section appears, which increases its diameter as dynamics progresses. It is observed in the dynamics that at t = 7.10x10^{-4} ms a bridge structure between the drops appears in the region of contact, which disappears at a later time due to the penetration of SPH particles between the drops. After a process of coalescence occurs (see figure 1 at t=7.70x10^{-4} ms), a bigger drop is formed (see this figure at t = 9.82x10^{-4}ms). Figure 2 shows the velocity vector field inside the droplets as well as in the region of contact between them at t=6.19x10^{-4}ms. It is important to see that inside the drops, the fluid tends to a velocity value around the initial velocity of 15.00 mm/ms, while in the area of contact between the drops an increase in the fluid velocity to a value of 26.0 mm/ms is observed. This increase in the fluid

velocity is due to changes on the pressure field inside the drops. In fact, in the region of contact of the droplets, the value of pressure reaches its maximum value.

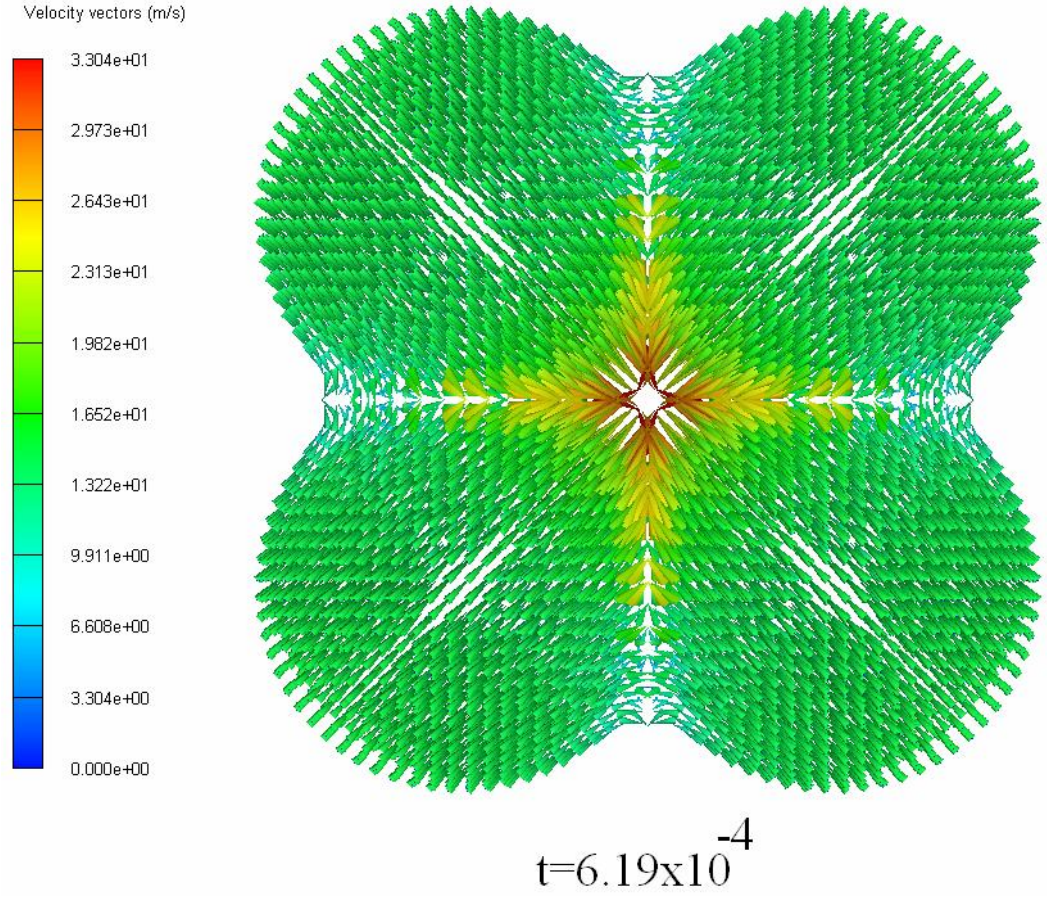

Figure 2. Velocity vector field for the collision between four drops at $t=6.19x10^{-4}$ ms (permanent coalescence process) with V_{col} = 15.0 mm/ms. The evolution time is given in milliseconds.

Another scenario for coalescence collisions is represented on figure 3. In this case droplets with a diameter of 30μm are defined inside the SPH code and each drop is composed by 5512 SPH particles, with a collision velocity V_x=15.0 mm/ms. In this case all velocities are directed to the center of the system with only one component of the velocity vector. It can be seen in figure 3 that at $t=2.14x10^{-4}$ ms a flat circular section appears and the drops that are placed at the center of the system have two flat sections. At $t=2.45x10^{-4}$ ms the coalescence process has begun and the particles of the drops start penetrating the region of the neighbor droplet. It can be seen at $t=5.04x10^{-4}$ms that the coalescence process is completed and the flat circular region between the drops disappears completely. Figure 4 shows the velocity vector field inside the droplets as well as in the region of contact between them at $t=2.14x10^{-4}$ms. As can be seen on figure 4, inside the drops the fluid tends to a velocity value around the initial velocity of 15.00 mm/ms, while in the area of contact between the drops an increase in the fluid velocity to a value of 26.00 mm/ms is observed.

On the other hand, it is observed that by choosing a collision velocity greater than the range of velocity values for the permanent coalescence, the phenomenon of fragmentation occurs. The following calculations were performed for droplets with 30μm of diameter: 5512 SPH particles for each drop, and a collision velocity of 30.0 mm/ms directed to the center of mass of the system. At the beginning of the calculation at $t=3.04x10^{-4}$ms the collision of the four droplets is shown in figure 5 and a flat circular section between the drops can be seen.

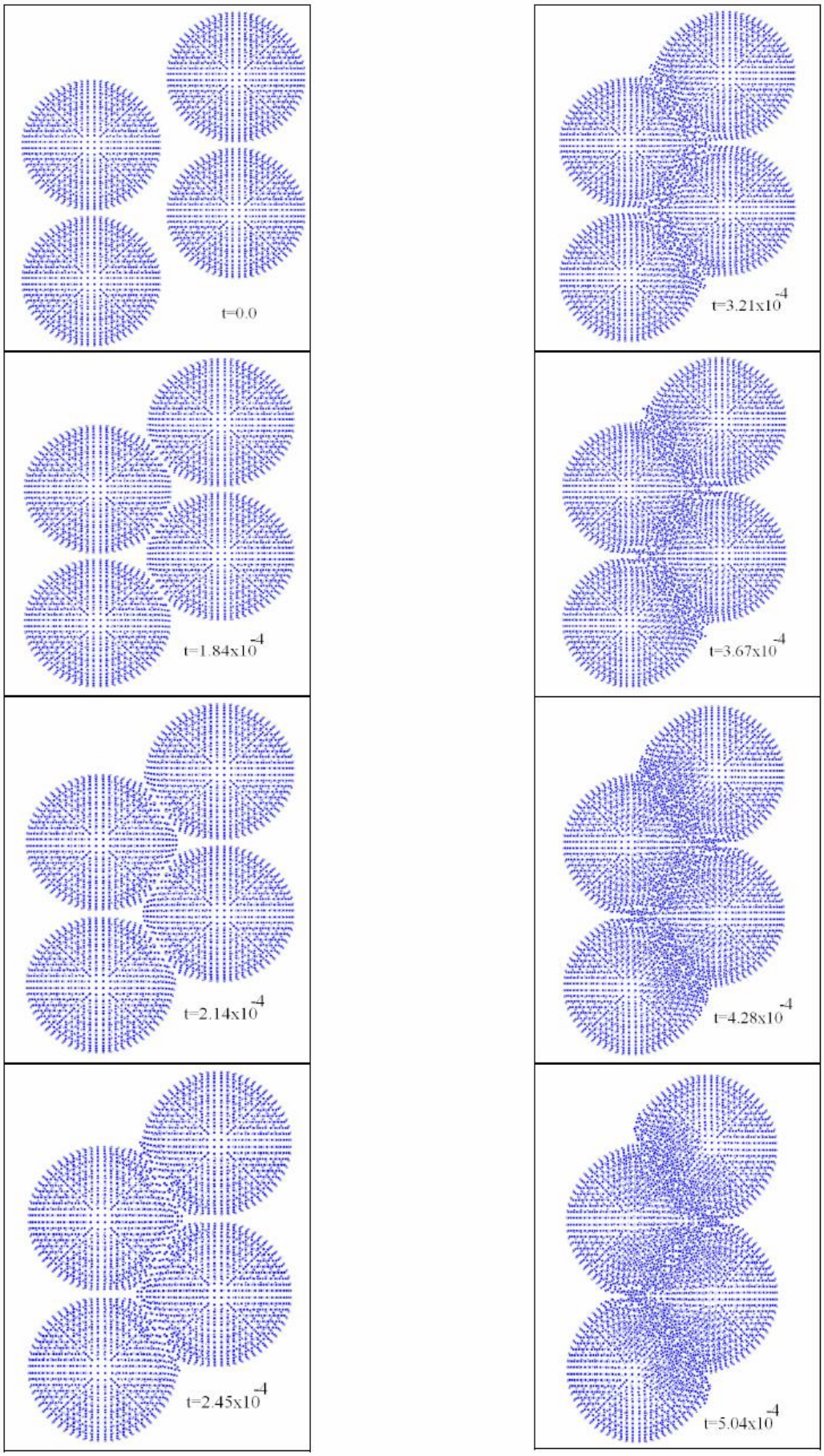

Figure 3. Sequence of times showing the evolution of the collision between four drops (permanent coalescence) with V_{col} = 15.0 mm/ms. The evolution of time is given in milliseconds.

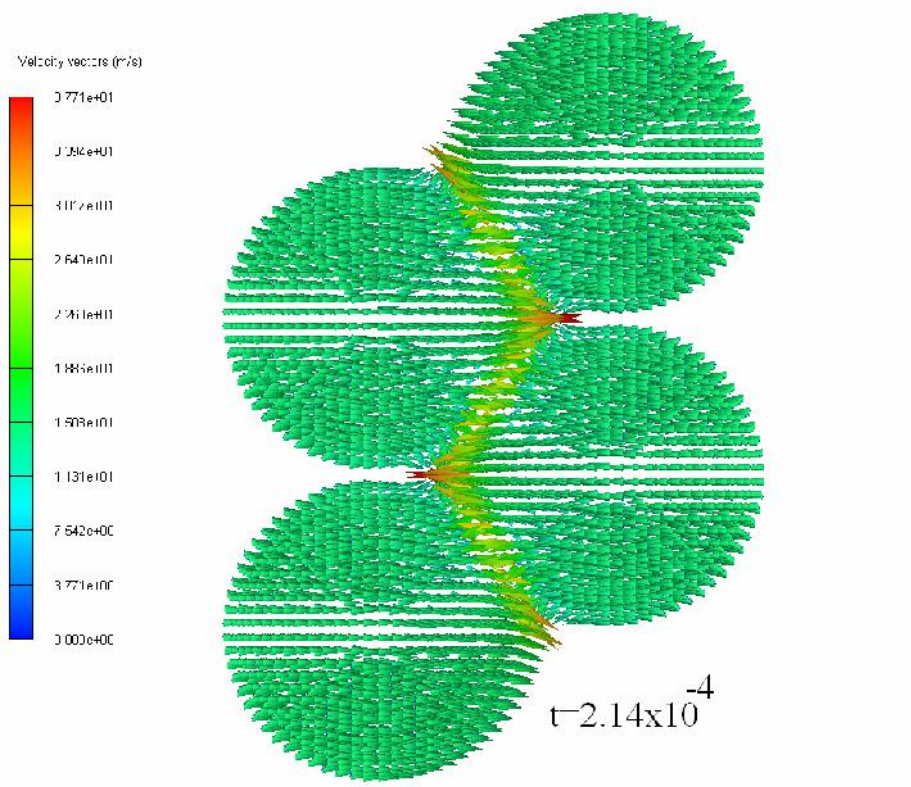

Figure 4. Velocity vector field for the collision between four drops at $t=2.14 \times 10^{-4}$ ms (permanent coalescence process) with V_{col} = 15.0 mm/ms. The evolution time is given in milliseconds.

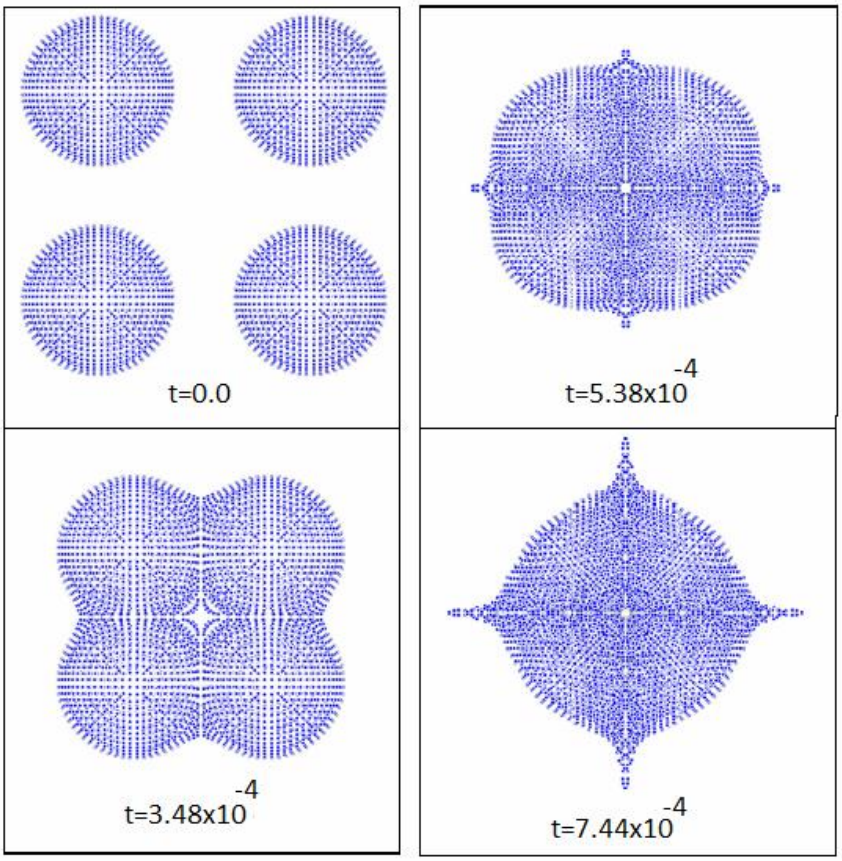

Figure 5. Sequence of times showing the evolution of the collision between four drops with V_{col} = 30.0 mm/ms. The evolution of time is given in milliseconds.

This section disappears completely at t =3.48x10^{-4}ms. It can be seen that at t=5.38x10^{-4}ms a portion of fluid that appears like a wave fronts, propagating in the plane x = 0 and y=0. These wave fronts begin to form little satellite drops at t=7.44x10^{-4}ms and these ligaments increment their amplitude as the dynamics runs. In fact these ligaments appear to be associated to an increase of satellite drops as well as to the fact that the bigger drop tends to take the shape of a spherical drop with four ligaments.

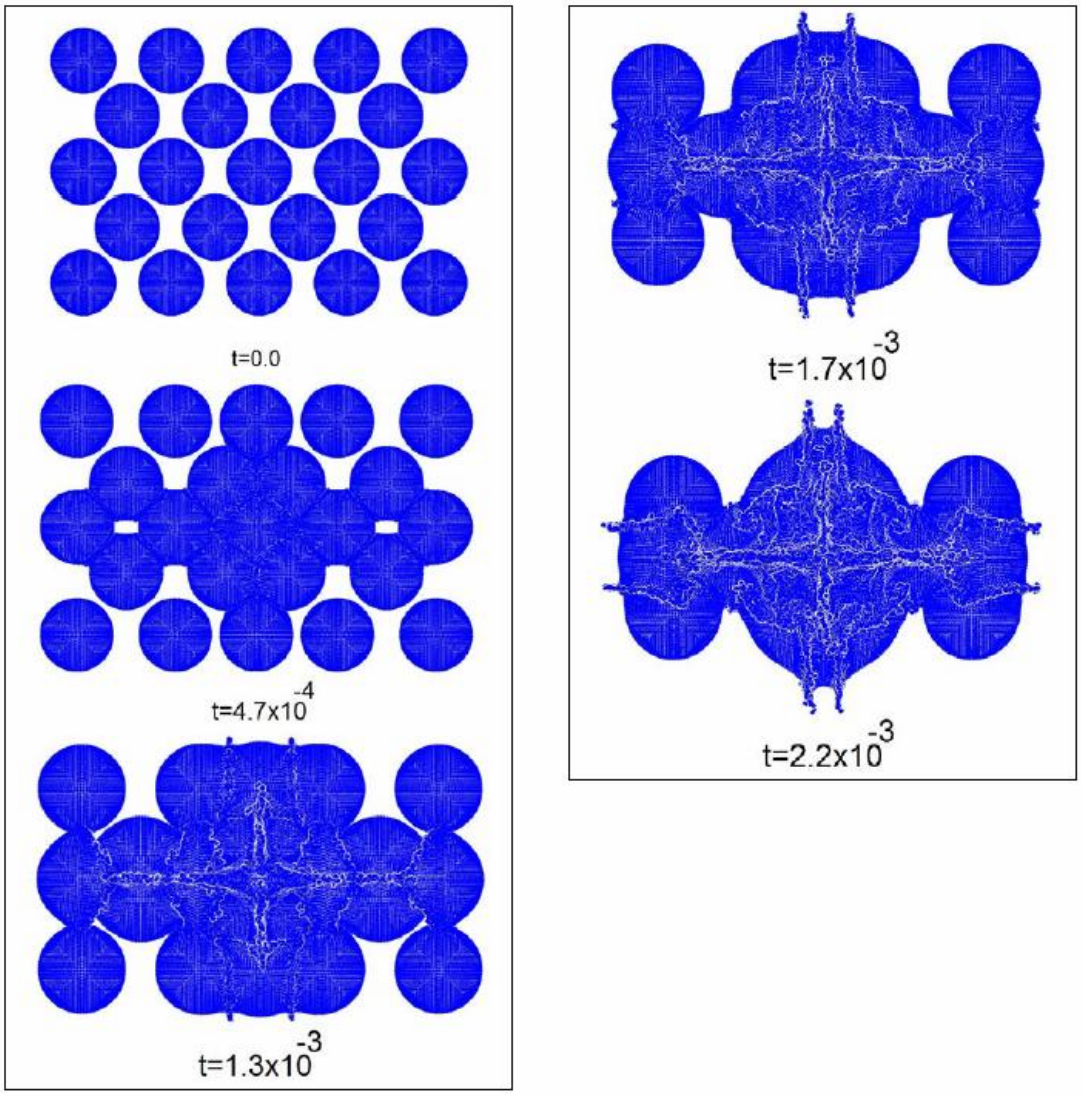

Figure 6. Sequence of times showing the evolution of the collision between the drops with V_{col} = 15.0 mm/ms. The evolution of time is given in milliseconds.

3.2 Coalescence, fragmentation and flocculation of clusters of drops

In order to model the collision of liquid drops, several calculations were carried out varying the velocity of collision for the modeling of the permanent coalescence of droplets in three dimensional spaces (3D) using the Smoothed Particle Hydrodynamics method. To do this, within the SPH code were defined 23 drops with diameter of 30μm and 15504 SPH particles for each drop (total number of particles: 356592) with a collision velocity of 15.0 mm/ms directed to the center of mass of the system.

It can be seen in figure 6 that at t=4.7x10^{-4} ms a flat circular section appears, which increases its diameter as dynamics progresses. It is observed in the dynamics that at t = 1.3x10^{-3} ms a bridge structure between the drops appears in the region of contact, which disappears at a later time due to the penetration of SPH particles between the drops. After a process of coalescence occurs (see figure 6 at t=1.7x10^{-3} ms), a bigger mass of liquid is formed (see this figure at t =2.2x10^{-3}ms).

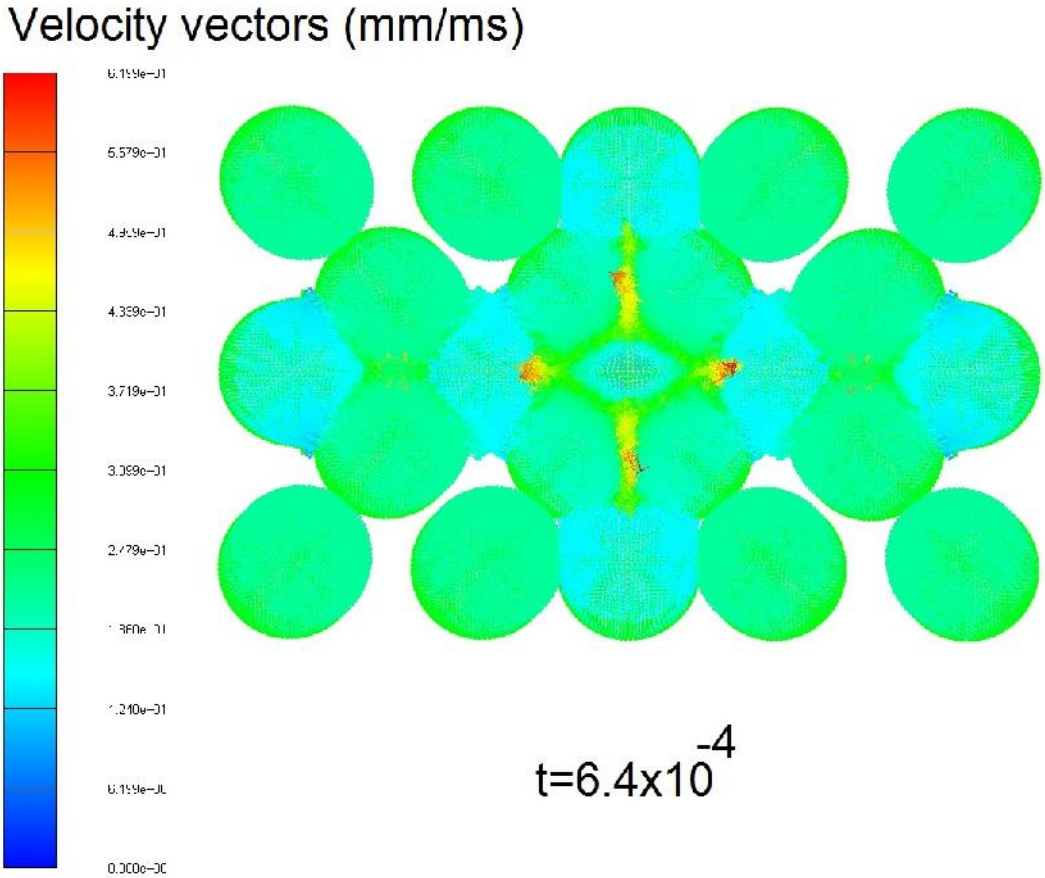

Figure 7. Velocity vector field for the collision between the drops at t=6.4x10^{-4} ms (permanent coalescence process) with V_{col} = 15.0 mm/ms. The evolution time is given in milliseconds.

Figure 7 shows the velocity vector field inside the droplets as well as in the region of contact between them at t=6.4x10^{-4}ms. It is important to see that inside the some drops, the fluid tends to a velocity value around the initial velocity of 15.00 mm/ms, while in the area of contact between the drops an increase in the fluid velocity to a value of 30.0 mm/ms is observed. This increase in the fluid velocity is due to changes on the pressure field inside the drops. In fact, in the region of contact of the droplets, the value of pressure reaches its maximum value.

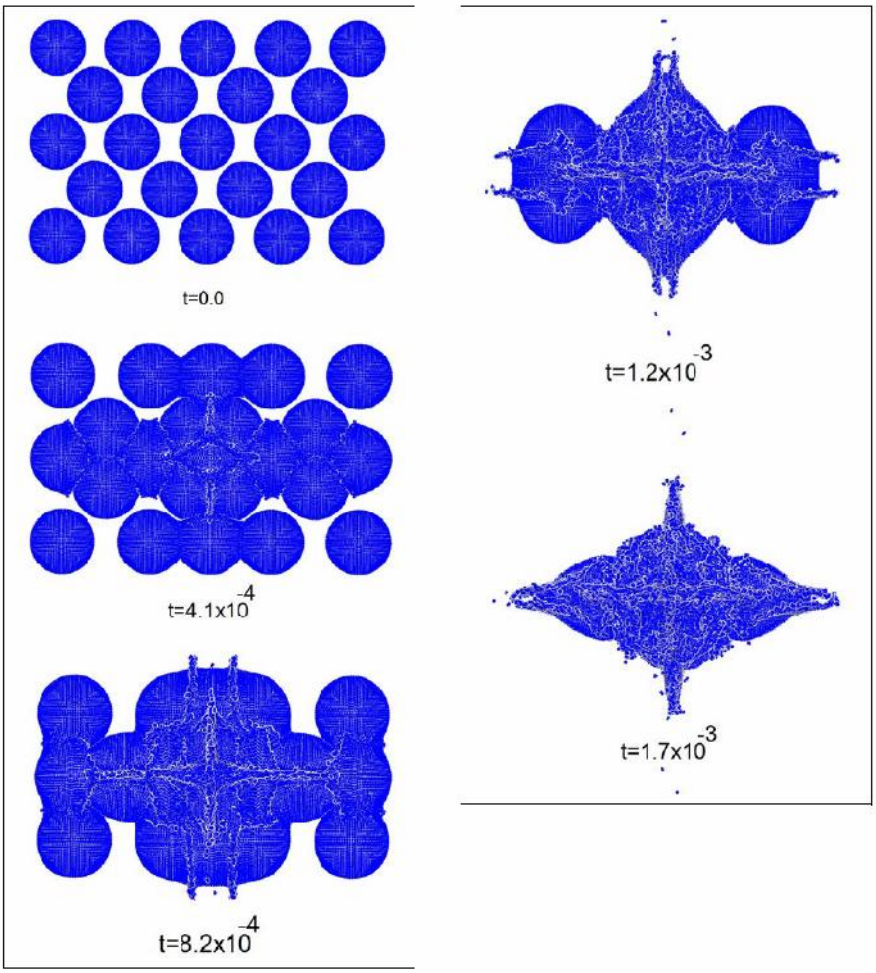

Figure 8. Sequence of times showing the evolution of the collision between the drops with V_{col} = 30.0 mm/ms. The evolution of time is given in milliseconds.

On the other hand, it is observed that by choosing a collision velocity greater than the range of velocity values for the permanent coalescence, the phenomenon of fragmentation occurs. The following calculations were performed for droplets with 30μm of diameter: 15504 SPH particles for each drop, and a collision velocity of 30.0 mm/ms directed to the center of mass of the system.

At the beginning of the calculation (see figure 8) at $t=4.1x10^{-4}$ms you can observe a flat circular section between the drops. This section disappears at $t=8.2x10^{-4}$ms and the coalescence of the drops begins. At $t=1.2x10^{-3}$ms a wave front arises from the resulting mass of fluid and this wave front start to form little satellite droplets. Figure 9 shows the velocity vector field just before the fragmentation of the drops has taken place. As shown in figure 9, the fluid velocity at the center of the some drops is around 30.00 mm/ms, while the fluid that is spread to the edges is accelerated, reaching a speed around 60.00 mm/ms.

Flocculation occurs when the collision velocity is decreased below the range corresponding to permanent coalescence. These calculations were performed for droplets having 30μm of diameter, 15504 SPH particles for each drop, and a collision velocity of 0.5 mm/ms. At the beginning of the calculation one observes at $t=6.7x10^{-3}$ms (see figure 10) that a flat circular section appears between the droplets, which at $t=1.1x10^{-2}$ms has already increased in diameter. As the dynamics runs a stretching of the surface of the drops until $t=1.1x10^{-2}$ms can be observed and the droplets form a floc. Figure 11 shows the velocity vector field at $t=1.1x10^{-2}$ms in the flocculation process. As shown in figure 11, the fluid velocity inside the some drops is around 0.50 mm/ms and at the zone of contact of the droplets the velocity is around 0.35 mm/ms.

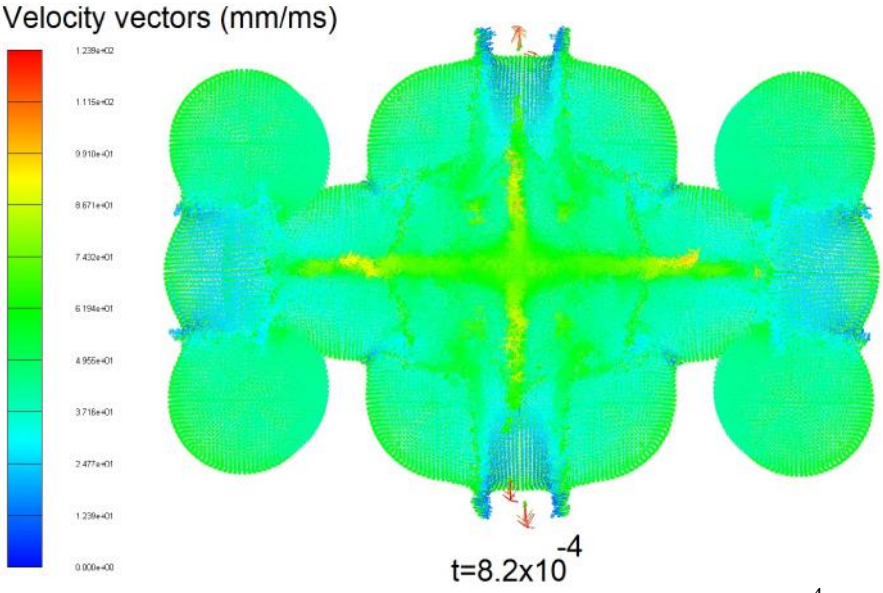

Figure 9. Velocity vector field for the collision between the drops at t=8.2x10^{-4} ms with V_{col} = 30.0 mm/ms. The evolution time is given in milliseconds.

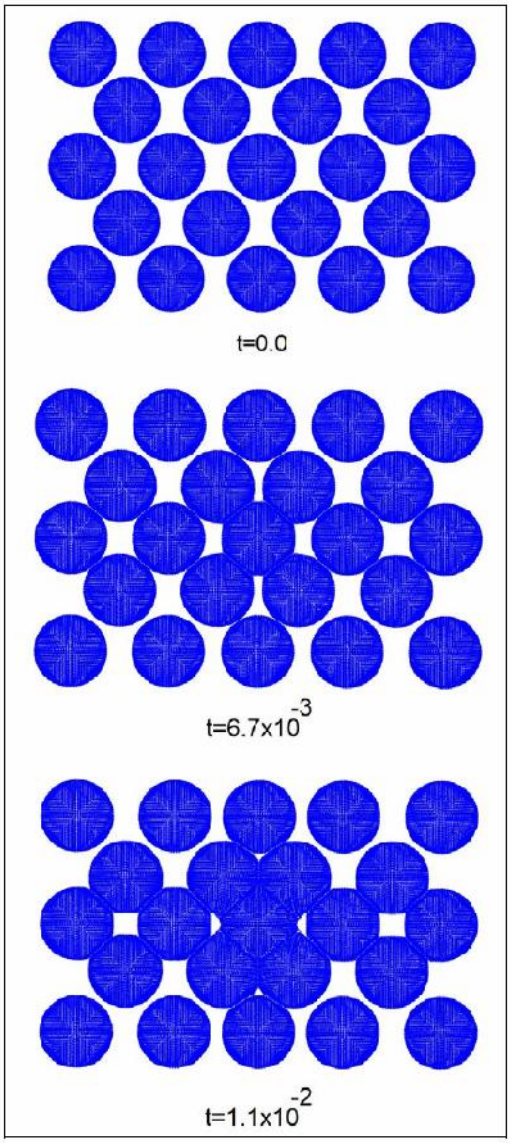

Figure 10. Sequence of times showing the evolution of the collision between the drops with V_{col} = 0.5 mm/ms. The evolution of time is given in milliseconds.

4. Conclusion

An adequate methodology has been proposed for the modeling of the multiple hydrodynamical collisions and formation of clusters of liquid drops using the SPH formalism in three-dimensional space. Several scenarios for multiple collisions of liquid drops in three dimensions have been carried out in our simulations. A flat circular section arises as the initial stage in all SPH calculations reported in this work. This flat circular section appears due to the existence of surface tension forces acting on each droplet. A range of values for the collision velocity is chosen and the possible outcomes (coalescence and fragmentation) for the collision process are obtained. The velocity vector fields were constructed and discussed for the different calculations. It can be seen that the fluid inside the drops tends to accelerate the SPH particles at the zone of contact between the droplets. This behavior is due to the non-uniform pressure differential inside the drops. At the zone of the drops that have no interaction with any other drop, the fluid tends to diminish the internal velocity. This can be explained by the behavior of the pressure field inside the drops, in fact, in that zone of the droplets, the inhomogeneous pressure field has a minimal value.

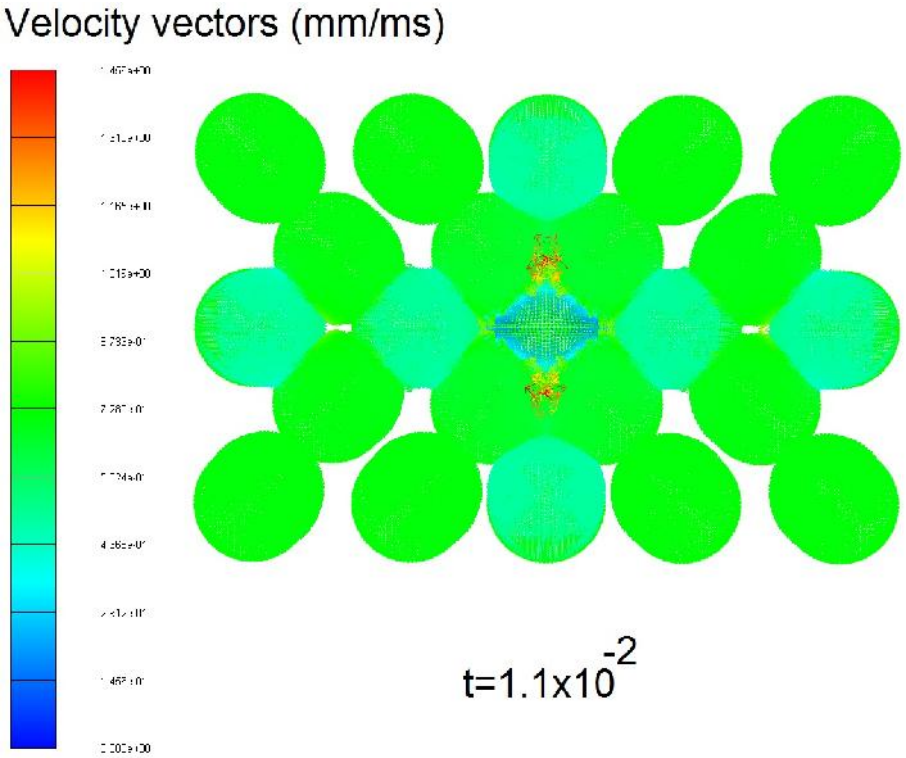

Figure 11. Velocity vector field for the collision between the drops at t=1.1x10^{-2} ms with V_{col} = 0.5 mm/ms. The evolution time is given in milliseconds.

REFERENCES

[1] P.S. Shah, L.T. Fan, I.C. Kao and L.E. Erickson, *Modeling of growth processes with two liquid phases: a review of drop phenomena, mixing, and growth.* Adv. Appl. Microbiol., 1972. **15**: p. 367-414.

[2] J.Y. Park and L.M. Blair, *The effect of coalescence on drop size distribution in an agitated liquid–liquid dispersion.* Chem. Eng. Sci., 1975. **30**: p. 1057-1064.

[3] P.J. Rourke and F.V. Bracco, *Modelling of drop interactions in thick sprays and a comparison with experiments, in: Stratified Charged Auto Engineering Conference.* Institute of Mechanical Engineering Publications, 1980. p. 101-116.

[4] S.G. Bradley and C.D. Stow, *Collision between liquid drops*. Philos. Trans. R. Soc. London Ser. A, 1978. **287**: p. 635-642.

[5] L. Roisman, *Dynamics of inertia dominated binary drops collisions.* Phys. Fluids, 2004. **16**: p. 3438-3449.

[6] Y. Yoon, F. Baldessari, H.D. Ceniceros and L.G. Leal, *Coalescence of two equal-sized deformable drops in an axisymmetric flow.* Phys. Fluids, 2007. **19**: p. 1-24.

[7] J. Eggers, J.R. Lister and H.A. Stone, *Coalescence of liquid drops.* J. Fluid Mech., 1999. **401**: p. 293-310.

[8] S.P. Decent, G. Sharpe, A.J. Shaw, and P.M. Suckling, *The formation of a liquid bridge during the coalescence of drops*. Int. J. Multi-phase Flow, 2006. **32**: p. 717-738.

[9] L. Rekvig and D. Frenkel, *Molecular simulations of droplet coalescence in oil/water/surfactant systems*. J. Chem. Phys., 2007. **127**: p. 1-16.

[10] F. Azizi and A.M. Al Taweel, *Algorithm for the accurate numerical solution of PBE for drop breakup and coalescence under high shear rates*. Chem. Eng. Sci., 2010. **65**: p. 6112-6127.

[11] X.Q. Xing, D.L. Butler, S.H. Ng, Z. Wang, S. Danyluk and C. Yang, *Simulation of droplet formation and coalescence using lattice Boltzmann-based single-phase model*. J. Colloid Interface Sci., 2007. **311**: p. 609-618.

[12] A. Acevedo-Malavé and M. García-Sucre, *3D Coalescence Collision of Liquid Drops using Smoothed Particle Hydrodynamics*. INTECH Publishers, 2011. **5**: p. 85-106.

[13] A. Acevedo-Malavé and M. García-Sucre, *Head-on binary collisions of unequal size liquid drops with smoothed particle hydrodynamics*. Research Signpost-Transworld Research Network, 2012. **9**: p. 245-266.

[14] A. Acevedo-Malavé and M. García-Sucre, *Coalescence collision of liquid drops I: Off-center collisions of equal-size drops*. AIP Advances, 2011. **1**: p. 1-17.

[15] A. Acevedo-Malavé and M. García-Sucre, *Coalescence collision of liquid drops II: Off-center collisions of unequal-size drops*. AIP Advances, 2011. **1**: p. 1-12.

[16] JJ. Monaghan, *Extrapolating B splines for interpolation*. J. Comput. Phys., 1985. **60**: p. 253-262.

[17] L.B. Lucy, *A numerical approach to the testing of the fission hypothesis*. Astron. J., 1977. **82**: p. 1013-1024.

[18] R.A. Gingold and JJ. Monaghan, *Smoothed particle hydrodynamics: theory and application to non-spherical stars.* Roy. Astronom. Soc., 1977. **181**: p. 375-389.