ILASS 2008
Sep. 8-10, 2008, Como Lake, Italy

Paper ID ILASS08-P-5

EXPERIMENTAL STUDY ON THE NUMBER AND SIZE OF MICROMETRIC DROPLETS PRODUCED BY THE IMPACT OF MILLIMETRIC DROPLETS ONTO A LIQUID FILM

C. Motzkus[1,2], F. Gensdarmes[1] and E. Gehin[2]

[1]Institut de Radioprotection et de Sûreté Nucléaire/Laboratoire de Physique et de Métrologie des Aérosols, B.P 68, 91192, Gif-sur-Yvette Cedex, France.

[2]Centre d'Etudes et de Recherches en Thermique, Environnement et Système, Université Paris XII, 61 av. du Général de Gaulle, 94010, Créteil Cedex, France.

charles.motzkus@univ-paris12.fr

ABSTRACT

The safety analyses of nuclear facilities require knowledge on the production of airborne particles in the case of dripping caused by leakages from a container. Indeed, during the impact of millimetric droplets onto a thin liquid film, secondary droplets with diameter below 50 µm are liable to be emitted and can evaporate rapidly in the air. If these droplets contain some radioactive solute, they give rise to airborne residues which are potential source of aerocontamination. Actually there is very few quantitative information in the literature on this subject. The purpose of this work is to study experimentally the droplets produced below 50 µm. An experimental setup was developed to measure the airborne particle mass and the size distribution of the aerosol produced during the impact of droplets, in a ventilated closed vessel. When a splash was observed, dry residues stemming from the evaporation of water droplets in the range 2-50 µm are measured.

1 INTRODUCTION

The safety analyses of nuclear facilities require extensive knowledge of accidental aerosol production in order to determine the potential sources of airborne contamination in the event of liquid falls such as dripping, caused by leakages from a pipe or a container. The study of the accidental generation of airborne radioactive contaminants in the nuclear field allows to estimate their potential effect on the operators, the facility and the environment. When millimetric droplets impact onto a thin liquid film, secondary droplets with diameter below 50 µm are likely to be emitted and can evaporate rapidly in the air. If these microdroplets contain some radioactive solute, they give rise to airborne residues which are potential sources of contamination. Currently, there is very few quantitative information in the literature on this subject.

In the case of millimetric water droplets, Okawa *et al.* [1], recently used their results and those of Stow and Stainer [2] to establish the relations determining on the one hand the number of the droplet produced with a diameter greater than 40 µm, and on the other hand the ratio of the total mass of secondary droplets to the mass of impacting droplets. Since there is a lack of knowledge about microdroplets production with a diameter less than 40 µm during the impact of millimetric droplets onto a liquid film, we are therefore explored this subject.

2 EXPERIMENTAL METHODS

2.1 Experimental setup and measuring methods

We have developed an experimental setup [3]. This setup allows us to measure the mass and the size distribution of the aerosol (airborne residues) produced during the droplets impacts onto a liquid film. The figure 1 presents a scheme of this setup. The experiments were performed in a closed vessel with a volume equal to 0.562 m^3 and a height of 86 cm. The closed vessel was ventilated with filtered air (HEPA - High Efficiency Particulate Air - filter).

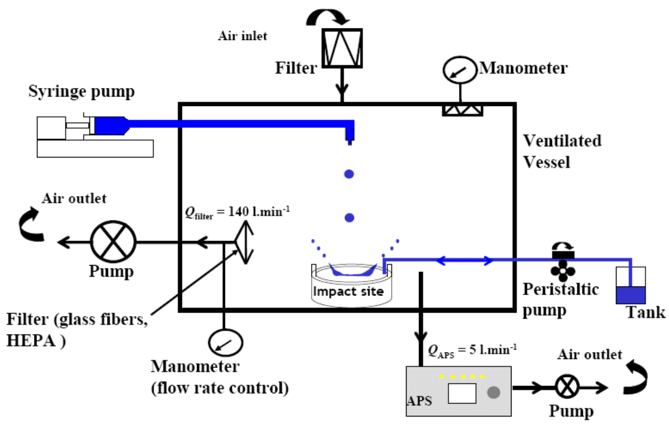

Figure 1. Scheme of the experimental setup [3].

Uniform water droplets with a diameter d_i of 3.9 ± 0.05 mm were produced using the hanging droplet method. Liquid is fed from a syringe to a hanging droplet on the tip of a needle until gravity forces exceed the forces due to surface tension and the droplet separates from the needle. The mean diameter of the droplets is determined by collecting a known number of droplets in a beaker and measuring the resulting mass of liquid. The one standard deviation, equal to be 0.025 mm, is obtained by multiple measurements. The temperatures of the liquid and the gas were equal to 20°C.

The impact velocity of the droplets is calculated from the height of their fall, taking into account the friction coefficient between the air and the droplet [4].

The impacting droplet creates a disturbance of the surface of the liquid film. The chosen time elapsing between two impacts (2.5 s) allows the surface to return to an undisturbed state (without apparent oscillation or wave). A stainless steel disk, with a low roughness (R = 0.18 µm), was suspended in a glass tank filled with the same solution as the one used for the droplets. The height of the submerged disk and the volume of liquid in the tank were adjusted to maintain a constant film thickness. The thickness of the liquid film was measured by means of a calibrated pins fixed on the disk.

We used a fluorescent tracer (sodium fluorescein) dissolved in the liquid in order to measure the mass of the airborne particles. The surface tension γ of this liquid is 66 mN/m and its viscosity μ_l is equal to 1 mPa.s. The density ρ_l is equal to 10^3 kg/m^3. Within the vessel the airborne particles are collected on a HEPA filter by means of a high flow rate sampler (cf. Fig.1). After each experiment, the mass of fluorescein collected is measured by placing the filter in a known volume of ammonia solution. The concentration of sodium fluorescein in the solution was then measured using a fluorimeter with a sensitivity of 10^{-11} g.cm^{-3}. It was decided that each experiment would last one hour, which was long enough to collect significant mass according to the sensitivity of the equipment used.

An Aerodynamic Particle Sizer (APS 3321-TSI) is used to measure the number concentration and size distribution of the airborne residues with aerodynamic diameters (d_{ae}) ranging from 0.5 to 20 µm. The flow rate of the incoming filtered air in the vessel is equal to the extraction flow rate Q (145± 3 L.min^{-1}), which is equal to the sum of the flow rates through the sampler and the APS.

Each experiment is composed of three phases. The first phase was the purging of the vessel with the sampling systems in operation. This phase lasted about 20 minutes. As soon as the concentration of particles measured by the APS became negligible, the experiment entered the second phase which constists in a dripping of droplets for one hour with an impact frequency f of 0.4 droplets per second (total of 1440 droplets). When the drip of droplets was stopped, the experiment entered the third sampling phase which lasted for a period of 20 minutes, long enough to collect all the airborne particles remaining in the vessel.

In parallel, the impact is observed using an ombroscopic method with a high-speed CCD camera (4000 frames / s and a spatial resolution of 25 µm / pixel). This device allows us to observe the phenomena like the prompt splash, delayed splash, deposition, etc.

2.2 Model used to characterize the microdroplets

We use a model [3] in order to define the relationships between the mass and granulometry of the aerosol sampled and those of microdroplets emitted during the impact. The model takes into account the particles settling during the sampling process. It is based on equations describing the evolution of the concentration of particles emitted in a closed vessel ventilated with filtered air. The concentration is supposed to be homogeneous in the vessel. We make the assumption that secondary droplets evaporate quickly and their settling during their evaporation period is negligible. The dry residues obtained are composed of fluorescein and have a density equal to 1500 kg.m^{-3}. These assumptions and the model were validated in a first study [3].

The equation 1 describes the increase of the concentration $C_1(d_{ae},t)$ during the second experimental phase, for a constant particles emission rate and an initial concentration equal to zero.

$$C_1(d_{ae},t) = \frac{q(d_{ae})}{A}\left(1 - \exp\left(-\frac{A}{V}t\right)\right), \tag{1}$$

where $A = v_{Set}(d_{ae})\ S + Q$.

$q(d_{ae})$ is the number particle emission rate. S is the settling surface area, in this case the floor of the vessel (0.6536 m^2). V is the volume of the vessel (0.562 m^3), v_{Set} is the airborne particle settling velocity and Q is the total volumic flowrate.

The equation 2 describes the decrease of the concentration $C_2(d_{ae},t)$ during the third experimental phase when the emission of particles has stopped at time t_1:

$$C_2(d_{ae},t) = C_1(d_{ae},t_1)\left(\exp\left(-\frac{A}{V}(t-t_1)\right)\right). \tag{2}$$

Using the equations 1 and 2, it is possible to express the mean concentration $C_{mean}(d_{ae})$ over both phases as a function of the particles emission rate:

$$C_{mean}(d_{ae}) = \frac{1}{t_2}\left(\int_0^{t_1} C_1(d_{ae},t)\, dt + \int_{t_1}^{t_2} C_2(d_{ae},t)\, dt\right). \tag{3}$$

Combining the equations 1, 2 and 3, it is possible to express the particle emission rate as a function of the mean concentration between $t = 0$ and t_2 as follows:

$$q(d_{ae}) = \frac{C_{mean}(d_{ae})\, A\, t_2}{\left(t_1 - \frac{V_e\left(1+e^{-\frac{A t_1}{V_e}}\right)}{A}\right) + \left(1 - e^{-\frac{A t_1}{V_e}}\right)\left(-\frac{A}{V_e}\left(1+e^{-\frac{A(t_2-t_1)}{V_e}}\right)\right)}. \tag{4}$$

In the case of aqueous solution of sodium fluorescein (concentration equal to 10 g/L), the mean number concentration $C_{mean}(d_{ae})$ is determined experimentally from the measurements of the mass and the normalised size distribution of the airborne particles, as follows:

$$C_{mean}(d_{ae}) = m \frac{6}{\pi\, d_{ev}^3\, \rho_{So}} \frac{F_v(d_{ev})}{V_s} \ , \quad (5)$$

where V_s is the volume sampled by the high flow rate sampler between t = 0 and t_2. m is the mass of sodium fluorescein collected on the HEPA filter and $F_v(d_{ev})$ is the normalised volume fraction of the size distribution of the dry residues as measured by the APS. ρ_{So} is the density of the particle.

The mean number of microdroplets of diameter d_s emitted per impact is obtained from the emission rate of the particles (Eq. 4) according to:

$$N^{impact}(d_s) = \frac{q(d_{ae})}{f} \ , \quad (6)$$

where f is the impact frequency (s^{-1}) and $q(d_{ae})$ is the emission rate of the particles with aerodynamic diameter d_{ae} resulting from the evaporation of the microdroplets (number of particles / s).

The diameters of emitted microdroplets (secondary droplets) are calculated from those of the dry residues by using the concentration of aqueous solution (10 g/L). The droplet with a diameter d_s are calculated with the equivalent volume diameter d_{ev} of the dry residue resulting from their evaporation:

$$d_s = \frac{d_{ev}}{\alpha^{1/3}} \ , \quad (7)$$

where α is the volume fraction of the non-volatile impurities in solution. In these experiments, this fraction is equivalent to the concentration of sodium fluorescein in solution. The relationship between the equivalent volume diameter d_{ev} and the aerodynamic diameter d_{ae} of a particle is given by the expression [5]:

$$\rho_{So}\, d_{ev}^2\, Cu\ (d_{ev}) = \rho_0\ \chi\ d_{ae}^2\ Cu\ (d_{ae}) \ , \quad (8)$$

where ρ_0 is the reference density of 1000 kg.m^{-3} and $Cu(d_{ae})$ is the Cunningham correction factor at the said diameter. χ is the dynamic shape factor of the particles and may be equal to 1 as the solid particles formed by the evaporation of a droplet of sodium fluorescein solution are assumed to be spherical.

3 RESULTS

When the splash was observed, dry residues stemming from the evaporation of droplets in the range 2-50 µm are detected.

For the impact of the aqueous fluorescein solution on a thin film (h = 1.2 mm) with a velocity v_i = 3.7 m.s^{-1} which corresponds to a Weber number We = 808 and an Ohnesorge number Oh = 2.0×10^{-3} the mean of several experiments give a total number of micrometric droplets emitted by impact N^{impact} equal to 104 with a standard deviation of 26.

Our results show that the count size distribution of the microdroplets emitted can be described by a lognormal function with a median diameter equal to 16 µm and a geometric standard deviation of 2 (fig. 2).

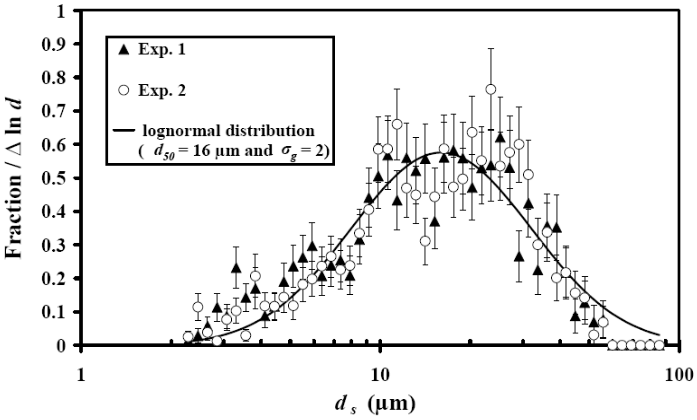

Figure 2. Count size distribution of the microdroplets emitted during the impact of a train of water droplets (h=1.2 mm, d_i = 3.9 mm; v_i =3.7 m.s^{-1}).

We suppose that these microdroplets may be produced on one hand by by the prompt splash (fig. 3) and on the other hand by the pinching of the fingers of the crown which occurs during the formation of the satellite droplets in the delayed splash (fig. 4). These phenomena are defined by Wander Wal *et al.* [6]: the prompt splash is associated with ejected droplets from the evolving rim concentric to the point of impingement or crown while it is still rising or advancing. In contrary, the delayed splash occurs at the stage of maximum expansion and it's associated with breakup of the crown.

These phenomena were observed with a high speed camera during the experiments.

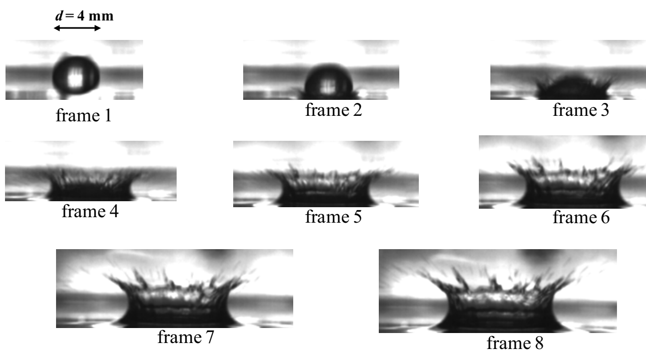

Figure 3. Photograph of the prompt splash during the impact of a water droplet on a liquid film (d_i=3.9 mm, v=3.70 m.s^{-1}). The time interval between frames is 0.25 ms.

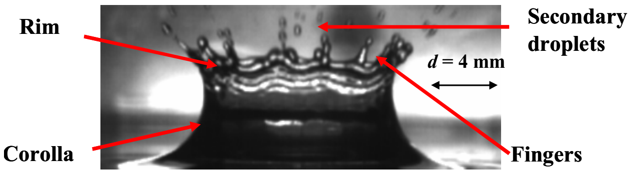

Figure 4. Photograph of the delayed splash 4.25 ms after the impact of a water droplet on a liquid film (frame 17, d_i = 3.9 mm, v_i = 3.70 m.s^{-1}).

4 CONCLUSION

This work describes the design of an experimental setup and the method used for the study of the emission of microdroplets during the impact of millimetric droplets onto a liquid film. The results obtained demonstrate the production of secondary microdroplets principaly with diameters less than 30 µm formed in the splash regime. Our results show that the count size distribution of the microdroplets emitted can be described by a lognormal function. In our experimental conditions, the median diameter of these microdroplets is around 16 µm. It is assumed that these microdroplets may be produced by two mechanisms: the prompt splash and the pinching of the fingers of the corona in the delayed splash. The continuation of this work is the study of the influence of key parameters (diameter and velocity of impacting droplet, viscosity and surface tension of the liquid and the thickness of the liquid film) on the production of microdroplets.

NOMENCLATURE

Symbol	Quantity	SI Unit
C	Concentration	Number / m^3
Cu	Cunningham correction factor	-
d_i	Diameter of the impacting droplet	m
d_s	Diameter of the secondary droplet	m
d_{ae}	Aerodynamic diameter	m
d_{ev}	Equivalent volume diameter	m
f	Impact frequency of droplets	s^{-1}
h	Film thickness	m
m	Mass of sodium fluorescein	kg
N^{impact}	Number of droplet emitted by impact	Number
Oh	Ohnesorge number ($= \mu_l / (d_i\, \gamma\, \rho_l)^{1/2}$)	-
q	Flow rate of particles	Number / s
Q_{ext}	Sampling volumic flow rate	m^3 / s
R	Roughness	m
S	Surface aera of floor	m^2
t	Time	s
v_i	Velocity of the impacting droplet	m / s
v_{set}	Airborne particle settling velocity	m / s
V	Vessel volume	m^3
V_s	Sampled volume	m^3
We	Weber number ($=\rho_l\, v_i^2\, d_i / \gamma$)	-
α	Volume fraction solute (sodium fluorescein)	-
γ	Surface tension	N / m
μ_l	Dynamic viscosity of the liquid	Pa s
ρ_o	Reference density	kg / m^3
ρ_l	Density of the liquid	kg / m^3
ρ_{So}	Density of sodium fluorescein	kg / m^3
σ_g	Geometric standard deviation	-
χ	Dynamic shape factor	-

REFERENCES

[1] T. Okawa, T. Shiraishi and T. Mori (2006). Production of secondary drops during the single water drop impact onto a plane water surface, *Experiments in Fluids*, vol. 41, pp. 965-974, 2006.

[2] C.D. Stow and R.D. Stainer, The physical products of a splashing water drop. *Journal of the Meteorological Society of Japan*, vol. 55, pp. 518-531,1977.

[3] C. Motzkus, E. Géhin and F. Gensdarmes, Study of airborne particles produced by normal impact of millimetric droplets onto a liquid film, *Experiments in Fluids,* Published online: 24 April 2008.

[4] K. Range and F. Feuillebois, Influence of surface roughness on liquid drop impact. *Journal of Colloid and Interface Science***,** vol. 203, pp. 16-30, 1998.

[5] P. Baron and K. Willeke, Aerosol measurement principles, techniques, and applications. New York: Wiley inter-science, 2001.

[6] R.L. Vander Wal, G.M. Berger and S.D. Mozes, Droplets splashing upon films of the same fluid of various depths. *Experiments in Fluids*, vol. 40, pp 33-52, 2006.