# A NOVEL ACOUSTO-ELECTRIC LEVITATOR FOR STUDIES OF DROP AND PARTICLE CLUSTERS AND ARRAYS

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## ABSTRACT

A novel and compact instrumentation for studying the behavior of drop sprays and of clusters of drops now permits fundamental research into the behavior of reacting and nonreacting fluid and solid species. The new capability is made possible by simultaneous acousto-electric levitation and charging of "seed" droplets (10-30 µm in diameter) which come together in 2-D clusters (with up to 300 droplets). These clusters are interesting in their own right because of their crystalline and quasi-crystalline forms, which depend on the acoustic and electric field parameters. By varying the electric and acoustic field intensities, one can cause a cluster of droplets to condense into larger drops (e.g. 50-300  $\mu m)$ which, because of their charge, form uniformly spaced 2-D arrays of monodispersed drops (e.g. 30-40 array drops in preliminary experiments). One or more layers of these 2-D arrays can form in the acoustic standing wave. Such a configuration permits a wide range of fundamental studies of drop evaporation, combustion, and nucleation. The drops can be single or multicomponent. Therefore, fundamental materials studies can also be performed. Using this same Cluster and Array Generation (CAG) instrumentation, it has been also possible in preliminary experiments to demonstrate the clustering and arraying of solid particles, both coated with an electrically conducting layer and uncoated, and both charged and uncharged.

# **INTRODUCTION**

The generation, control, and applications of sprays and aerosols cover a vast subject of relevance to the energy, environmental, and materials sciences. Fuels, soot and other environmental by-products, coatings, and intermediaries in particle synthesis all begin as sprays and aerosols. Understanding the behavior of multi-drop and particle systems and controlling the processes that created them require a good correspondence between theoretical modeling and benchmark experiments. The results of this characterization will provide benchmark data for comparison with theoretical analyses and numerical modeling, with the goal of developing an ability to predict changes in the process as we vary acoustical, electrical, and material parameters.

The literature on clusters and arrays of particles or drops (sprays) is very considerable and therefore is only selectively reviewed. Single drop evaporation and/or combustion has been studied with a wide variety of techniques, a few of which are reviewed here. Models of evaporation have been validated, for example, by levitating a drop either electrodynamically<sup>1</sup> or acoustically.<sup>2</sup> These techniques are also appropriate for multicomponent drops.<sup>3</sup> High

temperature liquid structure has been investigated using acroacoustic levitation techniques which permit the handling of heavy particles or drops.<sup>4</sup> Yet for many processes involving dense sprays, evaporation, ignition, and combustion processes are controlled by the interaction of droplets<sup>5</sup>; therefore, single drop evaporation models are entirely inadequate.

The study of the combustion of drops and sprays has advanced both experimentally and theoretically, as reviewed recently in the volume edited by Chiu and Chigier.<sup>6</sup> Some of the important advances illustrate how the evaporation, ignition and combustion of sprays are controlled by different parameters for dense vs. dilute clusters because of droplet interactions. Bellan, for example, has shown theoretically that for multicomponent drops, when the volatility of the solute is much greater than the solvent, liquid mass diffusion is important for dilute sprays, but not for dense sprays.<sup>7</sup>

"The allure of small controlled groups of droplets," according to Dunn-Rankin et al. "is that they provide an opportunity to isolate the effects of neighboring droplets on drop aerodynamics, drop vaporization, and drop combustion" which is not possible in spatially and temporally unsteady fields characteristic of a full spray or flame.<sup>8</sup> Their studies with streams has been one way to control the environment so that modeling and experimentation can be compared.

Annamalai has extensively surveyed the literature on the evaporation and combustion of arrays and clouds and has presented the PSI cell model as a way of determining relationships between group and spray combustion results.9 Annamalai concludes that "no detailed theory is available on flame spread involving an array of drops," and that cloud ignition and combustion data in an unconfined environment is lacking.<sup>11</sup> In his review, Chiu reviews the research accomplishments in droplet and spray combustion over a 40 year period. He concludes: "Needless to say, extensive experimental studies must be initiated to validate theoretical and numerical results."<sup>12</sup> In his report, Tambour considers the influence of multisize sprays on flame properties, spanning the range from small drops (20-25 µm), which are comparable to the "seed drops" we discuss below, to large drops (80-100 µm) which are of the same order as the array drops we produced with our apparatus.13

## Need for a benchmark system

These studies barely begin to scratch the surface of the broad amount of theoretical, experimental, and numerical work being done to understand drop and particle clusters and arrays, and their many applications. Just as the single drop became the central focus of very dilute evaporating or combusting systems, we need equivalent controllable benchmark multi-drop systems to validate dense spray evaporation and combustion models.

Control of arrays of drops and/or particles has recently improved with further development of the original work on multi-particle levitation using an electrodynamic balance by Wuerker et al.<sup>14</sup> The group of E. J. Davis, for example, used a double-ring electrodynamic balance apparatus to levitate NaNO3 particles with particle size nominally of 3.5  $\mu$ m.

(These were generated by producing solution drops of about 30  $\mu$ m with a vibrating orifice generator and passing them through a drying tube.) <sup>15</sup> They were able to achieve improved control of particle size and charge, as well as to manipulate the relative spacing of the particles. This novel approach permits a wide variety of experiments where the interaction of the particles or drops is crucial to the observed phenomena.

# CURRENT DEVELOPMENTS WITH CAG APPARATUS

Compact apparatus for levitating small drops is shown in Figure 1. It is an acoustic levitation cell driven by a composite transducer consisting of back-to-back piezoelectric discs, an aluminum horn, and a suitable reflector required for establishing an acoustic standing wave at approximately 28.2 kHz. If several drops of liquid are simultaneously levitated in this apparatus, acoustic radiation forces will normally drive them to a position just below a pressure node along the axis of the system. There they will aggregate and coalesce.



Figure 1: Levitation Apparatus

The top and bottom reflectors in the acoustic standing wave cell are plates of a capacitor that can be held at a potential varying from a few hundred to three thousand volts. The result of this new capability<sup>16</sup> is that a liquid placed on the lower plate can be atomized and charged by the vibrations of the electrodes. As the "seed" droplets are drawn toward the levitation plane, we note that they aggregate into two dimensional clusters (which we define to be groups of drops that are virtually touching). As seed drops are added to the cluster, there comes a point when the drops suddenly

coalesce into a single drop (with perhaps a few "petals" of seed drops on the exterior of the drop). This coalesced drop becomes part of a two dimensional array of drops, as the process is repeated with new seed drops and clusters. An array, for the purposes of this discussion, is characterized by drops spaced by more than one drop diameter. We are left with a number of important questions:

- What defines the size of a seed drop?
- What defines the size of the cluster of seed drops?
- What triggers the cluster to coalesce?

• How uniform in size are the array drops arising from cluster coalescence?

• What determines the spacing of the array drops and how uniform is this spacing?

• What determines the degree of stability of the array?

• If cluster size, array drop size, and array drop spacing could be controlled, as well as drop material and host gas, what applications would suggest themselves, both in terms of

validating theoretical models and in terms of technological significance?

• Can solid particles be similarly clustered and arrayed, and if so, what are the controlling parameters and potential applications?

# Initial observations and interpretation with the new apparatus

The fluid dynamical processes by which seed drops become clusters, and clusters become arrays have transient phases which are difficult to capture in still figures. We have put short video sequences of these processes on the World Wide Web at URL: www.yale.edu/bubble/array

#### Seed droplet formation

It is observed that if a few drops of ethanol are placed on the bottom plate of the 28 KHz resonator, then at a threshold level of vibration atomization, clustering, and arraying processes commence, leading to two or more planes of collected drops near the pressure nodes of the sound field (Fig. 2a, next page). These collected clusters and arrays have originated from a mist of fine droplets rising from the bottom plate of the cell (Fig. 2b). These drops, which we call "seed" drops, are approximately 25-40 µm in diameter and fairly uniform in size. The mechanism of generation is quite clear. The capillary wavelength of the ethanol surface is given by  $\lambda_c = (2\pi\gamma/\rho f^2)^{1/3}$ ; here  $\gamma$  is the surface tension,  $\rho$  is ethanol's density, and f is the acoustic frequency. According to Lang et al.<sup>17</sup>, the drop diameter from the surface instability ~ 0.67  $\lambda_c$ . In the present case, the observed size is somewhat smaller than the predictions. The discrepancy may be attributed to the state agitation of the surface or the small electric charge on each seed drop.



Figure 2: Droplet Cluster and Array Examples.

#### **Cluster formation**

Each seed drop is attracted toward the center of the cell by acoustic radiation forces as described in ref. 16. They aggregate together into a two dimensional cluster, as shown in the two-figure sequence 2c and 2d, but because of the small electrical charge on each drop, they do not immediately coalesce. The size of these clusters is determined by the balance of acoustic attraction and electrical repulsion. The acoustic attraction comes from two sources, as described in our lab's earlier work<sup>18</sup>: 1) the primary acoustic radiation force draws all drops to the same position in the sound field, and 2) a secondary, attractive interparticle force draws the drops toward each other. The repulsive force is simply coulombic in nature. The maximum size is determined when a new seed drop feels sufficient repulsion as to be expelled from the periphery of the flat cluster.

By increasing the acoustic field or decreasing the charge on the seeds, the cluster size can grow. Figure 2e shows a remarkable 2-D cluster consisting of 242 seed drops! Note also in Fig. 2f that under the right circumstances these clusters may undergo a fissioning process.

#### **Cluster coalescence**

The interparticle forces between drops plus the gentle agitation of the acoustic field increase the probability that two of the seed drops in the cluster will touch and coalesce, as shown in Fig. 2g. In the left part of the figure it is clear that the seed drops are beginning to form dimers and trimers

as a precursor to the rapid (10-50 mscc) formation of a cluster, which then coalesces to an array drop, as shown on the right side of the figure. The initial coalescence starts a chain reaction, because the secondary interparticle force goes as the diameter of the drop to the sixth power. As soon as two drops coalesce, increasing their effective diameter, the forces of attraction become stronger, and the process promotes more and more coalescence, until almost all of the drops have formed into a single drop, which we call an "array" drop for reasons to be made clear shortly. A "before" and "after" coalescence sequence is shown in Fig. 3; here 85 seed drops of about 45  $\mu$ m diameter have coalesced into a single drop of approximately 200  $\mu$ m in approximately 50 ms (which is determined primarily by the strength of the secondary interparticle acoustic forces.

Fig. 2g also shows the array drop with a few "petals," which are seed drops that haven't coalesced, perhaps because the smaller total drop area of the array drop (compared to the sum of the areas of the seed drops) increases the surface charge density, and thus produces greater coulombic repulsion.

# Array formation, and array drop charge

As each array drop forms, it is repelled from the center of the levitation cell by the newly generated seed drops which are aggregating and then coalescing. Each array drop remains in the potential well of the acoustic field but finds a new position with a minimum total energy of the system. This is a dynamic equilibrium, with the array of drops moving, and sometimes rotating around the central position. Fig. 2h shows a 2-D array of drops formed in the combined acoustical and electrical fields. The figure also shows a background pattern which look like triangles. This is just another plane of a drop array (out of focus), as the acoustic levitation cell can support two or three layers of drop arrays.

One can control the motion of the drop array by altering the acoustic and electric fields. For example, by lowering the acoustic field and raising the electric field, the primary force for levitation against gravity becomes electrical. In fact, this is precisely how the charge of the array drops can be measured. If the electric field is reduced too much, the drops will fall out. This is just the "Millikan oil drop experiment." In this way, we have observed that the drop charge density is approximately 1/1000 of the critical charge density —  $q^* =$ 

 $8\pi (\epsilon \gamma a^3)^{1/2}$ , which is referred to as the Rayleigh limit.

Figures 4 and 5 show the results of varying acoustic and electric field parameters. We see that as the electric field goes up for a constant acoustic field, the charge per seed drop goes up, and therefore the cluster size at which coalescence occurs decreases. Increased acoustic field strength for a constant electric field tends reduces drop spacing without having a major effect on drop size.



From these figures we note a few remarkable features

• The drop spacing appears uniform and controllable with electric and acoustic field adjustments.

• The array drop size seems reasonably uniform. We illustrate this uniformity by simply measuring the size of each drop using a CCD camera, frame capture, and a drop-size analysis system developed for the analysis of our space shuttle data.<sup>19</sup> Figure 6 gives two size distributions for two different sets of conditions, illustrating that at higher electric fields, the drop distribution is narrower and the mean drop size is smaller.

## Benchmark capability for drop clusters and array

The initial investigations reported above suggest that this new "Cluster and Array Generation," or CAG apparatus, can provide benchmark conditions for testing a significant set of questions related to evaporation and combustion studies. Moreover, the proper engineering of this capability may provide opportunities for new technologies in energy, pollution, and materials processing studies.



Figure 6: Drop size distributions for two different field conditions

# Important unanswered questions

The initial work focused primarily on one liquid, ethanol. Clearly, the liquid properties that will be important for single component liquids include: surface tension and density. For multiple component drops, we can consider a number of cases:

• The second component is a surfactant, which primarily affects the surface tension and the surface viscosity. Additionally, surfactants may provide a barrier to seed drop coalescence.

- The second component may be a dissolved species.
- The second component might be small solid particles.

The physical characteristics that will be important are: seed drop size and drop charge. Size is affected by surface tension, density, and more strongly by acoustic frequency. Charge is affected by the electric field. But after seed drop formation, the electric field and acoustic fields can be altered, taking on important roles in influencing final array drop size and spacing.

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