

This is an interesting paper on a fundamental application of the well-known cone-jet electro spray in materials science: the production of thin films of inorganic materials in critical technological areas such as batteries and fuel or solar cells, to name but a few. The morphologies and mechanical properties of these films are crucial for the performance of the devices. The authors carry out a fairly extensive survey of the literature on the subject, compiling a significant body of accumulated data in order to draw up targeted general “schedules” for obtaining the desired morphologies, based on the operating parameters of the electro spray (essentially the flow rate), which depend on the physical properties of the solutions of the precursors used to form the films.

The paper is well written and organized, and would probably be suitable for publication, but I wonder why the authors chose *Aerosol Research* rather than a more specific materials’ journal, given the approach and ultimate motivation of this paper. In any case, since the generation of an aerosol (more precisely, a fine liquid spray) is a necessary step to form the films objective of this work, it might fit in AR, but in this case I have a number of observations that the authors should address. I will focus on previous literature that I obviously know well:

- 1- The authors state (line 80) that previous electro spray studies do not provide a systematic way of optimizing different parameters to achieve the desired morphology. After reading the present manuscript, the reader is left with the impression that this work does not represent a drastic improvement on the situation that the authors claim in their statement on the existing literature (a statement with which I do not agree). In fact, while there is very precise evidence for the predictive power of droplet size and charge from well-established scaling laws, the authors use terms such as 'long', 'short', 'high' and 'low' (v.g. Table 1) without any correlation or comparison with precise quantities that could ultimately be reduced to non-dimensional parameters. In this respect, this paper is incremental, but not groundbreaking, because the authors' approach follows the same "cookbook" approach as many other works of this kind. This is not to say that this approach is not valid: on the contrary, it may be the only way to describe a precise know-how in a very complex parametric domain, but in this case the authors should tame the generality of their claims that would justify publication in AR.
- 2- Please give due credit to previous contributions, and recognize the historic sequence of events in relation to existing scaling laws for the emitted electric current and droplet size:
 - a. Expression (2) is the most consistent and accurate scaling law for the emitted electric current in a steady cone jet, but it appears to have been introduced prior to the work of Hartman et al. in 1999: Ganan-Calvo, Barrero and Pantano (1993, *J. Aerosol Sci.* 24, S19-S20) first presented this scaling law. Please mention this.
 - b. Ganan-Calvo (1997, *Phys. Rev. Lett.* 79, 217-220) introduced for the first time the scaling law (5)-(7) for the droplet size. Please see equation (15) in that work. Please mention this.
 - c. If the authors cite Hartman et al. 1999 in relation to (2) and (5)-(7), they must also cite Ganan-Calvo 1999, *J. Aerosol Sci.* 30, 863-872, who simultaneously introduced the same scaling laws in the same issue of the same journal as Hartman et al. 1999.
 - d. Ganan-Calvo et al. (1997, *J. Aerosol Sci.* 28, 249-275) devoted a section to the minimum flow rate and introduced a criterion similar to (10). Please mention this.
 - e. As far as I know, expression (11) was first introduced in Ganan-Calvo et al. 2013, *New J. Phys.* 15, 033035 (see equation (17) and figure 2 in that work).
 - f. In addition, the "minimum" flow rate for viscous fluids given by (12) has already been introduced by Montanero et al. 2011 (*Phys. Rev. E* 83, 036309, page 5, first paragraph) in the context of the minimum flow rate of flow focusing. Since this phenomenon is closely related to the Taylor cone jet electro spray, since they share a conical meniscus, a short note on the relevance of this expression is needed.

- g. Despite the previous point, the same work by Montanero et al. 2011, as well as Ganán-Calvo et al. 2013, discuss the other minima of the flow rate covering the whole parameter space for both Taylor cone jet and flow focusing.

Please mention all this relevant information and provide a general discussion if possible.

- 3- Lines 165-170: the droplet spreading on the surface depends on two critical parameters, in addition to the viscosity and surface tension: the droplet charge prior to impact, and both the liquid and the substrate conductivity. This also applies to the dried layer left by previous droplets on the substrate.
- 4- Lines 211-212: Due to its enormous width (many orders of magnitude), the conductivity range of a liquid cannot be described as "not too large" or "not enormously" influencing the droplet diameter in general. This is especially true when we are dealing with solutions exposed to extreme electric fields, whose solutes (e.g. salts) can induce local conductivity values many orders of magnitude different in the same liquid domain, depending on the intensity of the local electric field and local conditions (e.g. a boundary).
- 5- The authors should take a look at the work of Lopez-Herrera et al. 2023, J. Fluid Mech. 964, A19, and at least mention that electrokinetics is a fundamental aspect to consider in Taylor cone jets when dealing with relatively complex solutions such as inorganic salts in organic liquids, whose dissociation paths can be extremely complex.
- 6- Line 225: When the authors mention "an intermediate droplet size", ¿how is this intermediate point quantified? ¿How is it quantitatively related to other parameters such as temperature, concentration, substrate, liquid properties and final layer properties? In the same context (and this is related to the previous point 1), ¿is it possible to have a general guideline on the basis of rational relationships between crucial parameters? My guess is that it is indeed possible, with some effort. This would give the paper the relevance that one would expect from the strong claims made both in the abstract and in the conclusions.
- 7- The authors of the present work make a very important recommendation to the scientific community to disclose ALL related physical properties of the liquid solutions used in the published works, not only those that appear to be of interest to their authors. Again, this should be accompanied by a clear and practical demonstration of the importance of these properties in relation to the properties of the formed layers (the ultimate objective of all this) in quantifiable terms. For example, the authors give a nice guide to the role of the boiling point of the solvent and the excess temperature of the substrate. However, they do not provide any guidance on this excess in relation to the ambient saturation of solvent vapor as a result of the process, ambient pressure, or the thickness of the previous layer already deposited, among several other critical effects.
- 8- Lines 305-310: One of the crucial parameters to be considered in the electrospraying of complex liquids is the applied polarity. The mobilities of the anions and cations, which give the overall values of the local liquid conductivity, can be radically different, as can the local dissociation reactions, if the applied polarity is positive or negative. Again, please refer to Lopez-Herrera et al 2023 and mention this issue in relation to the present work.

I encourage the authors to carefully consider all of the above points. In summary, this interesting and ultimately valuable paper can be greatly improved by addressing these and other points and suggestions already made by other reviewers.