# Reply to comments by S. A. Ryce on "Charge-to-mass relationships for electrohydrodynamically sprayed liquid droplets"

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# Reply to comments by S. A. Ryce on "Charge-to-mass relationships for electrohydrodynamically sprayed liquid droplets"

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The "fissionability" concept developed by Ryce *et al.*[1–3] and applied to electrohydrodynamically sprayed liquid droplets provides a useful means of classifying various characteristics of electrohydrodynamic spraying phenomena. In particular, the concept permits the lumping together of many influences in the complex electrohydrodynamic spraying process and the classifying of complicated behavior in terms of a single fissionability parameter. This reply indicates that the fissionability concept complements the physical model developed by Pfeifer and Hendricks.[4] Additionally, attempts are made to add physical insights by correlating the fissionability coefficient ($y$) with physical parameters, notably the conductivity ($\sigma$), permittivity ($\epsilon$), kinematic viscosity ($\nu$), and density ($\rho$) of the working fluid and also with characteristics of the spraying such as the mass flow rate ($\dot{M}$), capillary diameter ($R$), droplet radius ($r$), and droplet mass ($m$).

In essence, the physical model developed by Pfeifer and Hendricks is directed toward a prediction of the distribution of sprayed particles about the minimum energy values originally computed by Vonnegut and Neubauer.[5] "Details of the spraying mechanism are not considered; the spraying phenomenon is modeled by the disruption of a portion of liquid with volume $V$ and charge $Q$ into $N$ particles."[4] Parametric studies of electrohydrodynamic spraying by Pfeifer and Hendricks[6] relate the charge-to-mass ratio of droplets to various characteristics of the working fluid and the spraying appratus. Only a region of the liquid surface at the capillary tip is presumed to become unstable in accord with the Rayleigh instability criterion[7]: "Studies involving the stability of fluid surfaces in the presence of intense electric fields indicate that disruption of the surface can be described as an exponential growth of surface perturbations···. Hence, the assumed state of dynamic equilibrium at the capillary tip is marked by the existence of an electric field within the dielectric; before enough charge accumulates on the surface to reduce such a field to zero, the surface becomes unstable and a charged droplet is ejected."[6] Thus, for many modes of electrohydrodynamic spraying, no parent drop really exists; the concept of a parent drop is merely a useful fiction in determining charge and mass relationships in an ensemble of droplets such that the ensemble energy tends toward a minimum state.

Some physical interpretation of the fissionability coefficient and its relation to a parent drop can be provided through analysis of certain characteristic times. Three time parameters characterize electrohydrodynamic spraying: the dielectric relaxation time $\epsilon/\sigma$, the viscous relaxation times $r^2/\nu$ and $R^2/\nu$, and the flow characteristic time $m/\dot{M}$. Three droplet formation regimes are discussed.

*Regime 1*, $\epsilon/\sigma \ll R^2/\nu$. As pointed out in the Comments by Ryce, very fine dispersions can occur for highly conducting fluids. Many of the photographic studies of electrohydrodynamic spraying in this regime indicate that, although the spraying may take on various forms, ejection of material occurs at one or perhaps several definite locales at the capillary tip. The parametric studies of electrohydrodynamic spraying of Pfeifer and Hendricks[6] utilize this fact in the development of their spraying model. Within this regime, the spraying may take on many forms dependent on the mass flow rate, applied voltage, and other parameters. For $\epsilon/\sigma \ll 2\pi R^3\rho/3\dot{M}$, the capillary is starved, and spraying typically occurs near the cylindrical edges of the capillary, sometimes at several points where irregularities of the capillary surface concentrate the local electric fields. As the mass flow rate increases, a hemispherical liquid interface develops at the capillary tip, and droplet ejection occurs at a tip of the interface. As the flow rate further increases, the material may be ejected into more and more elongated droplets until finally a jet of material is formed. Various pulsating modes may exist. Experimental evidence indicates that, with final dispersion into droplets, $m/\dot{M} \sim r^2/\nu$ (i.e., for glycerine with $r=1.5\times10^{-6}$ m, $m=9\times10^{-15}$ kg, $\dot{M}=5\times10^{-9}$ kg/sec, and $\nu=1.6\times10^{-2}$ stokes, $m/\dot{M}=1.8\times10^{-6}$ sec, while $r^2/\nu=1.4\times10^{-6}$ sec), and $\epsilon/\sigma > r^2/\nu$ (i.e., for glycerine with $\epsilon/\epsilon_0=42$ and $\sigma=5\times10^{-6}$ mho/m, $\epsilon/\sigma=80\times10^{-6}$ sec).[6] This regime corresponds to a fissionability $y>1$.

*Regime 2*, $\epsilon/\sigma \sim R^2/\nu$. Here the nature of droplet formation is strongly dominated by flow characteristic times. For $\epsilon/\sigma \ll 2\pi R^3\rho/3\dot{M}$ the spraying typically occurs as in regime 1 with the formation of small particles with relatively large charge-to-mass ratios (again, $m/\dot{M} \sim r^2/\nu$, $\epsilon/\sigma > r^2/\nu$, $y>1$). As the mass flow rate increases, the droplet size increases until $R^2/\nu \gg 2\pi R^3\rho/3\dot{M}$, where droplet formation is dominated by gravitational and surface tension effects. Asymmetric droplet formation and division may occur. This portion of the regime corresponds to a fissionability $y \leq 1$.

*Regime 3*, $\epsilon/\sigma \gg R^2/\nu$. For liquids which are very poor conductors, dispersion into droplets is relatively difficult. In some cases, for very pure materials, spraying due to charge conduction mechanisms may not occur at all. Generally, the spraying phenomenon is dominated by gravity and surface tension effects so that asymmetric droplet division occurs. This regime corresponds to a fissionability $y<1$.

In conclusion, the physical model developed by Pfeifer and Hendricks employs a hypothetical parent drop as an intermediate stage in the determination of the energy state of an ensemble of particles, assumes the Rayleigh instability criterion to be applied only to localized regions in the dynamic droplet production process, and relates charge-to-mass ratios to various physical parameters. The application of the fissionability concept to the hypothetical parent drop permits categorization of a wide range of electrohydrodynamic spraying phenomena independently of the details of spraying, and thus complements and extends the physical model.

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# Comments on "Statistical characteristics of Reynolds stress in a turbulent boundary layer"

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Gupta and Kaplan[1] present data on the moments of streamwise and vertical turbulent velocities ($u$ and $v$) and Reynolds stress ($uv$) in a laboratory turbulent boundary layer. They found that while $u$ and $v$ had flatness factors near the Gaussian value of 3, the $uv$ product had a flatness factor of about 10 which they judged to be large. We suggest that it is more natural to compare moments with the *joint* Gaussian predictions.

If $u$ and $v$ are jointly normal and each has a zero mean, one can calculate from the characteristic function[2] that

$$\begin{aligned}\overline{(uv)^2}&=\overline{u^2}\,\overline{v^2}(1+2\rho^2),\\ \overline{(uv)^3}&=(\overline{u^2})^{3/2}(\overline{v^2})^{3/2}(9\rho+6\rho^3),\\ \overline{(uv)^4}&=(\overline{u^2})^2(\overline{v^2})^2(9+72\rho^2+24\rho^4),\end{aligned}\tag{1}$$

where $\rho$ is the correlation coefficient $\overline{uv}/(u'v')$. The joint Gaussian predictions for the rms level, the skewness, and the flatness factor are then

$$\begin{aligned}\sigma&=[\overline{(uv)^2}]^{1/2}/u_*^2=(1+2\rho^2)^{1/2}/|\rho|,\\ S&=\overline{(uv)^3}/[\overline{(uv)^2}]^{3/2}=(9\rho+6\rho^3)/(1+2\rho^2)^{3/2},\\ F&=\overline{(uv)^4}/[\overline{(uv)^2}]^2=(9+72\rho^2+24\rho^4)/(1+2\rho^2)^2.\end{aligned}\tag{2}$$

In the region $50<y_*<100$, Gupta and Kaplan indicate that $\rho$ is in the range $-0.4$ to $-0.5$. Table I shows that their $\sigma$, $S$, and $F$ values agree fairly well with the joint Gaussian predictions.

Gupta and Kaplan found Reynolds number similarity for the first three $uv$ moments over the range $R_\theta=1900$–$6500$, with $R_\theta$ based on the free stream speed and momentum thickness. We can extend this range tremendously by introducing atmospheric results obtained in the log region between $y=5.66$ and 22.6 m in the neutral boundary layer over a homogeneous Kansas plain (further details of this experiment are given by Wyngaard *et al.*[3]). Length scales are so much larger in the atmosphere that $R_\theta$ is $10^4$–$10^5$ times the values of Gupta and Kaplan. Our $y_*$ values range from $10^4$ to $10^6$, and $\rho$ was $-0.3$. The first three $uv$ moments, which have uncertainty on the order of 10%, are compared in Table I. Note that the new data also agree fairly well with the joint Gaussian prediction.

The difference in the rms levels is due to the lower $\rho$ in the atmosphere, perhaps caused by very large scale

TABLE I. Comparison of stress moments.

| Parameter | Measured Value | | Joint Gaussian Value | | |
|---|---|---|---|---|---|
| | Gupta and Kaplan | Atmosphere | $\rho=-0.3$ | $\rho=-0.4$ | $\rho=-0.5$ |
| $\sigma$ | 2.2 | 3.2 | 3.6 | 2.9 | 2.4 |
| $S$ | −2 | −2 | −2.2 | −2.6 | −2.9 |
| $F$ | 10 | 10 | 11.3 | 12.1 | 12.7 |