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Dielectric Charged Drop Break – up at Sub-Rayleigh Limit Conditions

J. S. Shrimpton
Imperial College of Science, Technology and Medicine
Exhibition Road, South Kensington
London, SW7 2BX, UK

ABSTRACT

The maximum charge a drop may hold, for an electrically isolated, electrically conducting drop, in vacuum, is defined by the Rayleigh Limit. For spray plumes of electrically charged drops this condition is clearly not met due to the space charge field. We would like to simulate such spray plumes and to simulate drop break up within them, using stochastic methods. Since many simulated particles are required a dynamic drop stability analysis is clearly not computationally feasible. Based upon a static analysis, and a thorough review of the previous experimental data on charged drop stability, it is shown that for dielectric drops in the presence of significant electric fields, and particularly those within spray plumes, the maximum charge a drop may hold is less than the Rayleigh Limit. Typical values of stable drop charge of 70–80% of the conducting drop Rayleigh Limit are predicted, and this is supported by a majority of recent experimental work. We present an explanation of the sub-Rayleigh Limit drop fission within charged spray plumes for dielectric drops, based upon a static, rather than a dynamic analysis. This permits sub-Rayleigh Limit drop fission to be incorporated into stochastic particle simulations.

Index Terms — **Rayleigh Limit, Dielectric, charged drop.**

1 INTRODUCTION

THE maximum amount of charge Q_{ray} a droplet of radius r_d may hold is the Rayleigh Limit

$$Q_{ray} = 8\pi\sqrt{\gamma\epsilon_0 r_d^3} \qquad (1)$$

This is classically derived using perturbation methods [1]. Here γ is the surface tension coefficient and ϵ_0 is the permittivity of free space. It is a good estimate of the maximum level of charge a drop can accommodate providing the drop liquid is a perfect conductor, the continuum in which the drop resides is a vacuum, and there are no imposed external electric field disturbances. There have been many experimental studies that investigate the Rayleigh Limit and in what follows, we present what we consider to be a representative historical evolution of the results and their accuracy. Early, predominantly single drop studies essentially confirmed the limit, while later studies, and particularly those that make spray measurements seem to suggest that the maximum drop charge is below the Rayleigh Limit.

Doyle et al. [2] investigated the Rayleigh Limit by using a Millikan type force balance and correlated the potential gradient required to keep an evaporating charged drop between the plates with loss of charge at fission and measured the drop size at the end of the experiment using filter paper. They concluded that a charged drop lost approximately 30% of its charge. A similar approach was used by Abbas and Latham [3] with the additional information on drop diameter variation being recorded by observation, and the Rayleigh Limit was confirmed to an accuracy of 15%. More accurate work was presented by Schweizer and Hanson [4] who found good agreement with the Rayleigh Limit, with a scatter of about ±4%, and that the disruption process resulted in a 33% charge loss accompanied by a 5% mass loss.

More recently doubt has been expressed regarding the accuracy of the calibration of the equipment and it has been suggested by Taflin et al. [5] that the inherent errors could be as much as 10%. Taflin et al. [5] reported an experiment that for the first time allowed determination of the mass and charge loss associated with the explosion a more reliable and accurate way. The experimental apparatus is similar to that given in Doyle et al. [4], but in [5]

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optical resonance spectroscopy was used to continuously measure droplet size, to an accuracy of better than 1 part in 10^4. The disruption of droplets in the range 20 to 66 μm diameter resulted in a mass loss of approximately 1 to 2.3%, and a charge loss in the range 9.5 to 18%. All the droplets were observed to burst before reaching the Rayleigh Limit, the actual critical charge being scattered around 80% of the theoretical maximum. The mass loss results are consistent with most of the earlier investigators who all found little mass loss, with the exception of Abbas and Latham [4] who measured between 20 and 30% loss. It is notable that the measured critical charge for a particular liquid was similar, even for different sized droplets, and that it was different for each liquid which may suggest the critical charge depends in some way on the material properties. Measurements by Richardson et al. [6] for charged drop stability were made using an electrodynamic levitator. As in the previous instance by Taflin et al. [5], light scattering techniques were used to measure droplet size, which gave good accuracy. This set of experiments was carried out in a vacuum, however, which had not previously been attempted. Experiments were carried out with two liquids, firstly with dioctylphthalate and secondly with sulphuric acid. For the first time, a good conductor, sulphuric acid, was tested and the results obtained for this liquid are somewhat different. They are not directly relevant to this study which is concerned with the stability of dielectric liquids, although they do highlight the dependence of material properties on the break up process. The droplets were held in a quadropole trap, the radius was monitored by an accurate light scattering method, and the charge was determined by periodic weight balancing. The oil droplets were found to lose approximately 15% of their charge and 2.25% of their mass on bursting, independent of their original size. Gomez and Tang [7] carried out experiments with heptane, including an additive to increase electrical conductivity, in ambient air and at atmospheric pressure. Here phase Doppler anemometry was used to measure the size of the droplets, although it is a less accurate technique than that used by Taflin et al. [5] still gives size information to an accuracy of 1 part in 10^2. In other respects, however, this experiment was slightly different to the preceding examples in that the droplets were sprayed from a capillary and mean stable drop charge was found to exist between 70 and 80% of the Rayleigh Limit. Significantly, an external field in the spray where disruption occurs is several orders of magnitude greater than that applied in levitation experiments with single droplets. Fernandez de la Mora [8] reports that using mean flow and current are poor indicators of mean drop charge due to the formation of satellite drops via either primary or secondary atomization mechanisms within the electrospray. With this in mind, the mean charge of drops can be thought of as accurate at best, or an overestimate at worst, since the charge in the satellite drops is considered negligible by Gomez and Tang [7]. Further work [9] shows the drop charge broadly varies with the cube of diameter, and that the drop charge of the large drops varies from 98 down to 55% of the Rayleigh Limit, decreasing with increase in flow rate. Clearly for the larger flow rates, coulomb explosions were occurring, to the extent that the satellites carry up to 30% of the charge.

The findings of the above work are summarized, in chronological order in Table 1. We can assume that more recent experiments give the most accurate results, largely due to the more advanced methods employed for measuring the size of the drops as the experiment progresses. In general, the recent, accurate experimental results show that break-up occurs before and not at the Rayleigh Limit. When break-up does occur, then mass in the range 1 to 5% is ejected from the parent drop, carrying a charge approximately 15% that of the parent. Most of the experimental evidence is mutually supporting, with a few notable exceptions. Abbas and Latham [3] were alone in reporting a much larger mass loss, and Richardson et al. [6] were alone in testing a conducting liquid, which showed a much larger but more random charge loss.

Thus far, we have not commented on how the charged drop breaks up once it has attained a critical charge level. Firstly, we may note that the charge may be ‘real’ or ‘induced’. ‘Real’ charge is free charge present on the drop surface. ‘Induced’ charge, relevant for dielectric drops, is polarization charge created by an electric field passing through the surface. Clearly, for electrically charged drops of dielectric liquids, in external electric fields there will exist both ‘real’ and ‘induced’ charge in the drop surface. Considering first uncharged dielectric (ferroelectric) drops in the presence of electric (magnetic) fields as investigated by Sherwood et al. [10] and Stone et al. [11]. The break up mechanism is a function of the dieletric (conductivity) ratios between the two fluids, e.g. ϵ_d/ϵ_g (σ_d/σ_d) and liquid viscosity. Considering henceforth solely dielectric liquids and electric field interactions, where the permittivity ratio is very large, $\epsilon_d/\epsilon_g \rightarrow \infty$ and approximating a perfectly conducting drop in a vacuum, then conical points form, as predicted by Taylor [12], and charge and mass may issue from this point. When the conductivities are finite, ie there exists an electric field inside the drop, then the drop still deforms. Providing $\epsilon_d/\epsilon_g \geq 20$ conical points still form, though the apex is different from the ‘Taylor-Cone’ angle of 49.3°. Below these permittivity ratios conical points do not form, and instead the drop extends along the field direction to form a cylindrical shape. Similar behavior may be observed when free charge is present on the drop surface [8] by applying scaling laws for electrosprays, formed from Taylor Cones and Cone-jets [13] to drops. Here, ‘polar’ and ‘non-polar’ models are required to explain the electrospray behavior, and in a similar manner to the shape deformation of uncharged drops of Sherwood [10] and Stone et al. [11], very non-polar liquids cannot form electrosprays.

Table 1. Summary of experimental results.

		Droplet Immediately Before break-up					Results		
Source	Procedure	Liquid	γ^* mN/m	ϵ_r^*	d (μm)	$\frac{Q_0}{Q_{Ray}}$	$\frac{Q_1}{Q_0}$	$\frac{M_1}{M_0}$	n
[2]	Single drop levitation and final ink spot calibration	Aniline	42.12	7.06	60 – 200	1	~ 0.7	~ 1	1 – 10
		Water	71.99	80.1					
[3]	Single drop levitation and regular aerodynamic balancing	Water	71.99	80.1	60 – 400	1	0.75	0.75	
		Aniline	42.12	7.06					
		Toluene	27.93	2.379					
[4]	Single drop levitation and ac spring voltage plate	n-octanol	27.10	10.3	15 – 40	1	0.77	0.95	
[5]	Single drop electrodynamic balance and optical resonance spectroscopy	Bromododecane	30.5	4.07	44	0.715	0.78		
		Dibromooctane	34.0	7.43	33	0.857	0.84	0.98	
		Dibutyl phthalate	34.09	6.58	20	0.747			
		Hexadecane	27.44	2.05	42	0.737	0.83	0.98	
		Heptadecane	27.9	2.06	33	0.795	0.88	0.98	
[6]	Single drop electrodynamic levitator and light scattering	Dioctyl phthalate	28.3	5.22	1 – 8	1.02	0.85	0.975	
		Sulphuric Acid	55.4	84		0.84	0.506	0.999	
[7]	Electrospray and phase Doppler anemometry	Heptane and 0.3% Stadis 450	18.1	1.93	– 5 130	0.6 – 0.8			~ 15
[9]	Electrospray and DMA	Benzyl alcohol and Dibutyl sebacate	33.9	6	~ 1		0.98 – 0.55		

*Data sourced from the experimental papers given in column 1 or Lide [14]

To summarize we notice analogous behavior between uncharged dielectrics in the presence of electric fields and charged dielectrics, with self-generated electric fields, and suggest that within charged spray plumes both space charge generated electric fields and externally applied electric fields effect the stability of dielectric charged drops. This is beyond the assumptions of the Rayleigh limit, which applies only to a pure conductor, with no external field disturbances.

2 MODEL DERIVATION

By noting the similarity in behavior between drops with electric charge in the absence of electric fields, and uncharged drops in the presence of fields, we seek to establish the magnitude of charge required to destabilize a given drop in a given electric field. Because of the fluid motion occurring inside dielectric drops [10] driven by the surface fields a single dynamic drop analysis would require a significant computational effort. We therefore analyze this net charge at the surface using a simplistic static approach and show how limiting solutions match the dynamic analysis. A perfectly conducting fluid droplet has an infinite supply of charge that can move within it, and so there will never be a steady state internal electric field. This is the situation to which the Rayleigh Limit applies, but it is not true for a dielectric, but charged, drop. In the liquids used in the fission experiments listed in Table 1 the permittivity was finite and low, so an electric field may have existed inside the drops due to the presence of the external field, originating from either the levitating field in the case of single drop experiments, or from the potential applied to the capillary, for electrosprays. This causes polarization of the drop fluid due to the realignment of the molecules in the liquid with the electric field. As a result, a charge is induced on the boundary of the liquid and this is shown diagrammatically in Figure 1, where on the right hand boundary the positive ends of the molecules are aligned, and similarly the negative ends of the molecules on the left hand boundary. It is important to note that the induced charge increases the total surface charge on one side of the drop and simultaneously reduces it on the opposite side. In terms of drop stability this means that one side of the drop is always more unstable than the same drop with no electric field present. Conversely, the opposite side is always more stable, though this of no interest in the present investigation. Overall however, the drop is

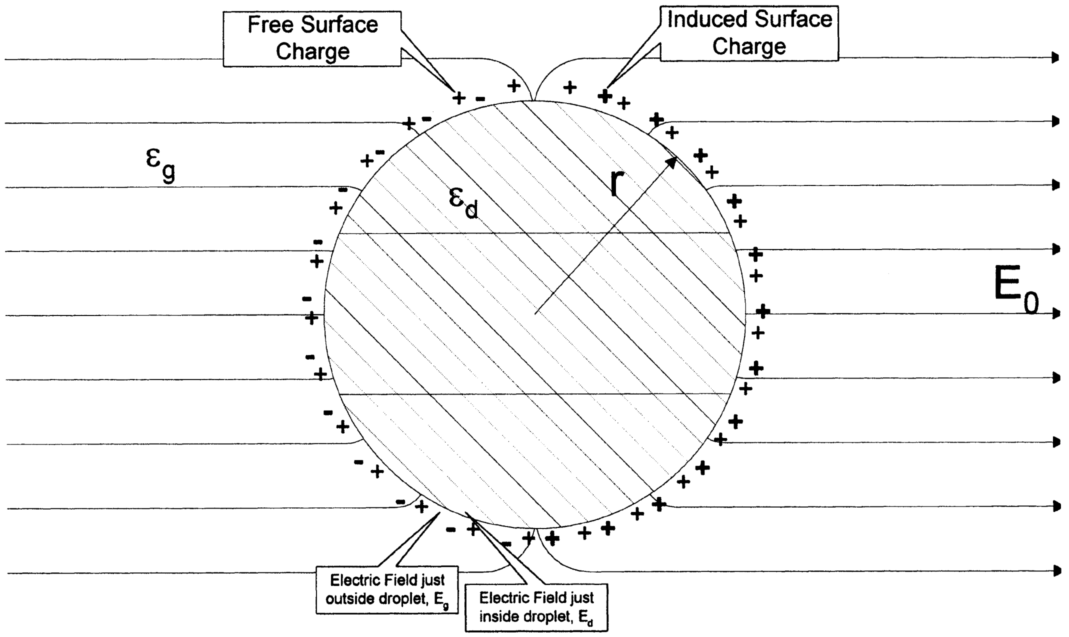

Figure 1. Drop break up nomenclature.

always more unstable, and the location of the point of maximum surface charge density is, as will be shown below, is a function of the sign of the free charge and the direction of the external electric field. It is proposed that this induced surface charge, Q_i, can contribute, with the free charge on the surface, Q_f, towards the fission of the droplet. This principle leads to the simplistic expression for the critical free charge on the droplet:

$$Q_{ray} = Q_f + Q_i \tag{2}$$

We are effectively stating that the drop critical condition is still the Rayleigh Limit, but that the induced charge, due to the dielectric nature of the liquid and the applied electric field, may contribute. This ensures that for a non-zero induced charge, the amount of free charge that may be carried is less than that predicted by the Rayleigh Limit. We also show that this approach, for zero free charge, correlates with the induced charge necessary for the disruption of uncharged drops in the presence of external electric fields. We find, Appendix A, that at the drop surface the induced charge density is given as

$$\rho_i = E_g \epsilon_0 \left(\frac{\epsilon_g - \epsilon_d}{\epsilon_d} \right) + \rho_f \left(\frac{\epsilon_d - 1}{\epsilon_d} \right) \tag{3}$$

where ϵ_d and ϵ_g denote the relative permittivity of the drop and continuum respectively. We assert that the electric field outside the drop, at and normal to the surface, E_g comprises elements due to the external field and the free surface charge. The electric field at the outer surface of an uncharged drop, in a uniform field E_o is given by

$$E_g = -3E_0 \left(\frac{\epsilon_d}{\epsilon_d + 2\epsilon_g} \right) \cos\theta \tag{4}$$

Now, considering the same drop, holding a charge Q_f, but with no external field

$$E_g = \frac{\rho_f}{\epsilon_g \epsilon_0} \tag{5}$$

Here we have assumed that the free charge Q_f is distributed evenly over the surface and thus does not generate an electric field inside the drop. Since equations (4) and (5) are solutions to the Laplace equation, they may be superimposed, and then substituting for the total E_g in equation (3) give

$$\rho_i = \rho_f \left(\frac{\epsilon_g - 1}{\epsilon_g} \right) + 3E_0 \epsilon_0 \left(\frac{\epsilon_d - \epsilon_g}{\epsilon_d + 2\epsilon_g} \right) \cos\theta \tag{6}$$

Equation (6) gives the surface charge density around the drop surface in terms of θ for a given externally applied field E_0 and free charge density ρ_f. The maximum will depend on the relative sign of ρ_f and the complementary sign of cos θ. For $\rho_f > 0$, the maximum charge density occurs when cos θ = 1, i.e.

$$\rho_i = \rho_f \left(\frac{\epsilon_g - 1}{\epsilon_g} \right) + 3E_0 \epsilon_0 \left(\frac{\epsilon_d - \epsilon_g}{\epsilon_d + 2\epsilon_g} \right) \tag{7}$$

Substituting into equation (2) gives the following result which predicts the maximum free surface charge a dielectric droplet in an electric field can hold

$$\frac{Q_f}{Q_{ray}} = \left(\frac{\epsilon_g}{2\epsilon_g - 1} \right) \left\{ 1 - \frac{3}{2} E_0 \sqrt{\frac{r\epsilon_0}{\epsilon_g \gamma}} \left(\frac{\epsilon_d - \epsilon_g}{\epsilon_d + 2\epsilon_g} \right) \right\} \tag{8}$$

We see from equation (8) that as $E_0 \to 0$, $\epsilon_g \to 1$, and as $\epsilon_d \to \infty$ then the Rayleigh stability limit is regenerated and as such the new relationship is well bounded. We now explore the behavior of equation (8) against published experimental data and comment on the results.

From the experimental work of Inculet and Kromann [14], if an uncharged droplet is placed in a sufficiently strong electric field, the induced charge will be sufficient to cause break-up. Using equation (8), the electric field strength to cause fission with zero free surface charge can also be obtained

$$E_0 = \frac{2}{3}\sqrt{\frac{\epsilon_d \gamma}{\epsilon_0 r}}\left(\frac{\epsilon_d + 2\epsilon_g}{\epsilon_d - \epsilon_g}\right) \tag{9}$$

From experimental observations, Taylor [12] suggests that for an uncharged drop in a uniform field, at the point of disintegration, the following correlation seems to hold

$$E_0\sqrt{\frac{r}{\gamma}} = C \tag{10}$$

where C is a constant.

By comparison with equation (9), the present work allows an expression for this constant to be obtained

$$C = \frac{2}{3}\sqrt{\frac{\epsilon_g}{\epsilon_0}}\left(\frac{\epsilon_d + 2\epsilon_g}{\epsilon_d - \epsilon_g}\right) \tag{11}$$

For a water droplet in oil, Inculet and Kromann [14] presented the experimental value of this constant as 1.91 $\times 10^5$ $(V/\sqrt{N})$. By applying (8), for a water drop in oil, with permittivities estimated as $\epsilon_d = 50$ and oil with $\epsilon_g = 2.2$, then $C = 3.74 \times 10^5$ $(V/\sqrt{N})$. Although these values differ by a factor of two the agreement is still thought reasonable since from the photographic evidence presented in Inculet and Kromann [14] the drop is highly non-spherical and thus our assumptions are not well approximated. We must also remember that we are using a static model, and thus charge motion is not considered, for shape evolution methods see Sherwood [10] and Stone et al. [11].

We also show how the modified Rayleigh Limit behaves as a function of E_0, the externally applied electric field, r, the drop radius, and ϵ_d, the drop permittivity in Figures 2 and 3. In these figures, the contours are the amount of free (i.e. real) charge, expressed as a ratio of the Rayleigh Limit (i.e. equation (1)) that is required to precipitate break-up. In Figure 2, which considers a drop of 40 μm radius, for $E_0 = 0$ V/m we recover equation (1), but for non-zero external fields the induced charge at the surface reduces the charge required, i.e. reducing the stability of the drop, to the extent that at large fields, the drop charge at break-up may only be 60-80% of the Rayleigh charge, which it is recalled is based on a pure conductor, in vacuum, with no external field. In Figure 3 we vary the drop radius for a low permittivity drop, typical of an insulating liquid, for instance a hydrocarbon. Again, large charged drops are highly unstable, unlikely to ever attain their Rayleigh charges, however small drops are little influenced. Inspection of equation (7) shows the reason to be that drop charge varies with $r^{3/2}$ whereas the induced charge varies as r^2. The location of break up will always be where the surface tangent is normal to the direction of E_0, and which side depends on the relative signs of Q_f and E_0. This behavior is observed experimentally in the photographic evidence provided by Gomez and Tang [7] where all drop disruption occurs away from the spray axis. The approximate stability limits as a function of E_0 and r also seem to be well predicted and we assert that charged drop stability in spray plumes is well represented by the modified Rayleigh Limit expression we propose. In particular, the detailed work of de Juan and Fernandez de la

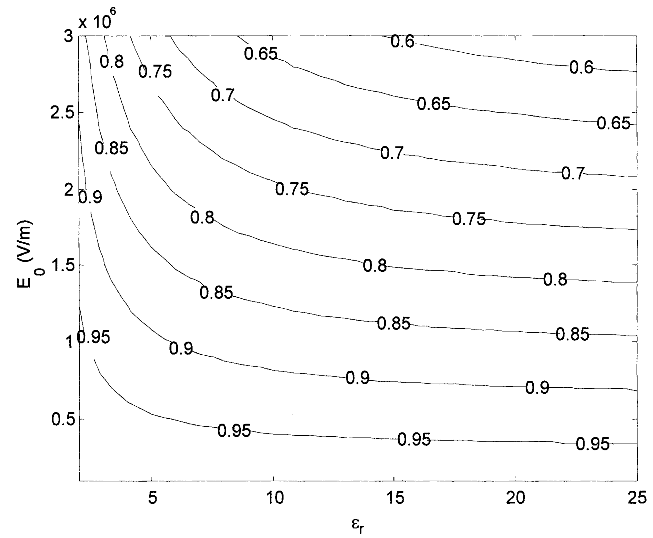

Figure 2. Distribution of the free charge fraction Q_f/Q_{ray} for r = 40 μm drop, where γ = 0.03N/m, ϵ_g = 1 as a function of electric field, E_0 and drop permittivity, ϵ_d.

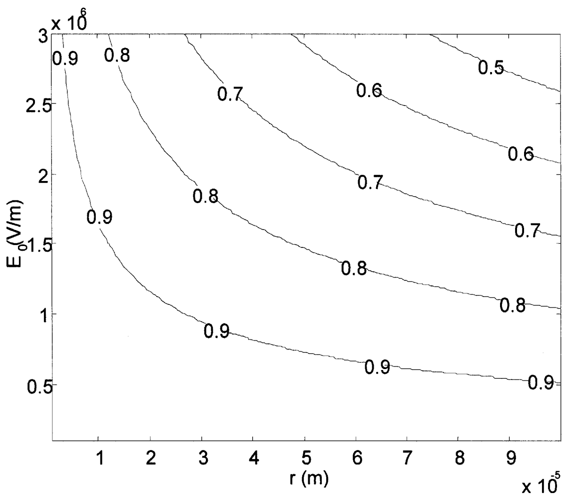

Figure 3. Distribution of the free charge fraction Q_f/Q_{ray} drop of permittivity ϵ_d = 2, where γ = 0.03N/m, ϵ_g = 1 as a function of electric field, E_0 and drop radius, r.

Mora [9] and Gomez and Tang [7] are well explained by using this model. In the latter paper estimates of electric fields inside the spray plume are available.

3 CONCLUSIONS

THE experimental literature concerning the measurement of the Rayleigh Limit has been reviewed from a historical perspective. We have shown that there is a trend for the more recent work, which uses more accurate investigative methods, to show that the Rayleigh limit is not obeyed in practice, but that single charged drops usually break up at between 70% to 80% of this limit and charged drops within spray plumes down to 55% of the Rayleigh Limit. We have proposed an extension to the Rayleigh Limit that incorporates effects due to the permittivity of the drop and the medium in which it resides. By assuming the free and induced charge in the drop surface do not interact, we have also included the destabilizing effect a linear external electric field has upon drop stability. Results from the proposed correlation show good agreement with published experimental data for charged sprays. The method proposed is analytical, and hence extremely computationally efficient. This permits the prediction of Rayleigh induced drop breakup in the presence of electric fields in stochastic spray simulations.

APPENDIX A

A.1 DERIVATION OF EQUATION (3)

The relationship between the displacement, $\underline{D}$, and electric, $\underline{E}$ and polarization $\underline{P}$ vectors in a linear dielectric may be written

$$\underline{D} = \epsilon_0 \underline{E} + \underline{P} = \epsilon \underline{E}\ ,\ \underline{P} = (\epsilon - \epsilon_0)\underline{E} = (\epsilon_r - 1)\epsilon_0 \underline{E} \quad \text{(A1)}$$

Considering the geometry of Figure A1
In each medium,

$$\underline{P}_1 = \underline{E}_1(\epsilon_0\epsilon_1 - \epsilon_0)\ ,\ \underline{P}_2 = \underline{E}_2(\epsilon_0\epsilon_2 - \epsilon_0) \quad (A2)$$

Since the polarization is equivalent to the surface charge at the interfaces, then the net surface charge at the left hand interface of the dielectric

$$\rho_i = \underline{P}_1 - \underline{P}_2 = \underline{E}_1\epsilon_0(\epsilon_1 - 1) - \underline{E}_2\epsilon_0(\epsilon_2 - 1) \quad \text{(A3)}$$

Any free charge at the interface defines any difference in the displacement fields of the two media

$$\rho_f = \underline{D}_1 - \underline{D}_2 = \epsilon_0\epsilon_1\underline{E}_1 - \epsilon_0\epsilon_2\underline{E}_2 \quad \text{(A4)}$$

Eliminating E_2 from equations (A3) and (A4) yields, at the left side of the dielectric

$$\rho_i = E_1\epsilon_0\left(\frac{\epsilon_1 - \epsilon_2}{\epsilon_2}\right) + \rho_f\left(\frac{\epsilon_2 - 1}{\epsilon_2}\right) \quad \text{(A5)}$$

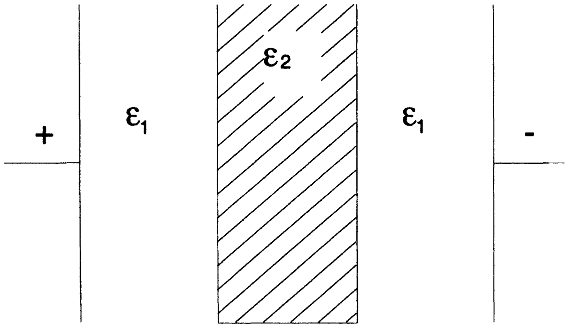

Figure A1. Structure of two dielectrics.

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John Shrimpton was born in November, 1968. He received the Ph.D. degree in mechanical engineering from UMIST, UK in 1995. In 1998, he gained a Lectureship in Mechanical Engineering at Imperial College, London and in 2003 was awarded an EPSRC advanced fellowship. His research interest continues to be the fluid dynamics of multiphase flow, with particular emphasis on those that contain electric charge.