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THE EFFECT OF ELECTRIC FIELDS ON MASS TRANSFER FROM FALLING DROPS

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Abstract—Studies of the effect of electric fields on mass transfer have been largely confined to liquid–liquid systems. In this investigation, the effect of applied potential gradients in the range 0–3·5 kV/cm on the rate of evaporation of acetone from drops of a 50 per cent acetone–water solution into a counter-current nitrogen stream have been measured. Data on drop size, charge, drop velocity and residence time are reported and it is concluded that increases in rates of mass transfer, by up to 50 per cent, are due to increased Reynolds' numbers rather than enhanced interfacial turbulence and internal circulation.

NOMENCLATURE

A,	drop surface area [cm^2];
D_N,	nozzle diameter [cm];
D_V,	diffusivity [cm^2/s];
d,	drop diameter [cm];
d_e,	equivalent spherical drop diameter on a volume basis [cm];
h,	electrode spacing [cm];
k_g,	mass-transfer coefficient [cm/s];
M,	mass transfer, (m/y_A) [mg];
M_w,	molecular weight [kg/kmol];
m,	amount of evaporation per drop [mg];
P,	total pressure [kN/m^2];
R,	universal gas constant [kJ/kmol K];
Re,	Reynolds' number $(d_e u\rho/\mu)$;
Sc,	Schmidt number $(\mu/\rho D_v)$;
Sh,	Sherwood number $(k_g d_e/D_v)$;
T,	temperature [K];
t,	drop fall time [s];
u,	drop velocity [cm/s];
V,	electric potential [kV];
y_A,	mole fraction of acetone vapour above liquid.

Greek symbols

γ,	interfacial tension [N/cm];
σ,	charge density [C/cm^2];
ε,	dielectric constant;
ε',	permittivity of free space [C/kV cm];
μ,	viscosity [Ns/cm^2];
ρ,	density [g/cm^3].

Subscripts

a,	average value;
c,	referring to a charged drop;
0,	referring to zero potential.

INTRODUCTION

WHILST the effects of electric fields on the hydrodynamic behaviour of liquid droplets are relatively well understood, the somewhat limited number of investigations into mass transfer in such systems have been largely confined to liquid–liquid systems. The importance of mass transfer from evaporating droplets into a gas phase, particularly in spray drying applications, has prompted this investigation into the effect of electric fields on such an operation. Previous work on the application of an electric field to drops formed at a nozzle before falling through the field has shown that three effects are apparent. Firstly the size of the drop is decreased, thus increasing the effective interfacial area per unit volume of liquid; secondly, the terminal velocity of the drop falling through the field is increased and finally internal circulation and interfacial turbulence are increased.

Considering these factors in turn, many workers have investigated drop deformation, particularly Harkins and Brown [1], Allan and Mason [2] and Stewart and Thornton [3] who modified the interfacial tension for the case of a charged drop to give:

$$\gamma_c = \gamma - (5\sigma^2 d_e)/(4\varepsilon\varepsilon'), \qquad \text{N/cm.} \qquad (1)$$

Thus the effect of increasing the electric field is to decrease the effective interfacial tension leading to a smaller drop and a larger number of drops forming

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at the nozzle for a given volume of liquid. Hendricks *et al.* [4] have shown that there are two distinct regimes of droplet formation, the induced charge regime and the electrostatic dispersion regime in which very small charged droplets are produced having a large specific charge. With high fields and low conductivity liquids, the spraying occurs as a series of irregular sprays around the periphery of the nozzle with little liquid moving along the axis of the nozzle. This effect was noted previously by Vonnegut and Neubauer [5]. A large number of workers [2,4–14] have studied the stability and distortion of liquid drops in electric fields and Stewart [15] has produced a correlation relating the induced charge to drop size, nozzle size and the physical properties of the system, though it was not possible to incorporate the applied field strength because the potential distribution between two electrodes immersed in an insulating liquid cannot be obtained from Laplace's equation $\nabla V^2 = 0$. Gray [16] has shown that this is due to accumulation of space charge near the electrodes.

Several workers [3, 15, 17, 18] have investigated the effects of electric fields on droplet velocities in liquid–liquid systems and it has been shown [3] that the terminal velocity of water drops in *n*-heptane can be almost doubled by the application of a field. Stewart [15] has found that in some cases however, the velocity reaches a maximum value; the group $(D_N^2\,\Delta\rho g/\gamma)$ being the controlling factor. In liquid–gas systems, a series of important investigations into the charge and velocity of monodisperse fuel droplets in air have been reported by Thong and Weinberg [19], and Jones and Thong [20]. In this work, an approximate theory is developed which correctly predicts the specific charge acquired by the droplets as a function of the applied field strength and this coupled with data on drop size, allows the mobility and hence the trajectories of drops to be predicted for any set of conditions. The classical work of Rayleigh [21] indicates that the charge required by a drop causes a reduction in oscillation frequency, though Stewart [15] has suggested that the oscillation frequency of a charged drop is no different to that of a charged drop although the acquired charge is likely to promote interfacial turbulence. Considering internal circulation, Taylor [10] has shown theoretically that the presence of a field enhances the internal flow of a drop and this has been confirmed experimentally by McEwan and De Jong [22].

It seems likely that these effects, increased interfacial area, drop velocity, internal circulation and interfacial turbulence, should enhance mass transfer from drops and yet the application of an electric field has received only limited attention; all reported investigations being confined to liquid–liquid systems. Thornton [6] has reported almost double the rate of extraction of benzoic acid from toluene, using water as the dispersed phase, with an applied field strength of 3·25 kV/cm. This work incorporated a counter-current flow, whilst with a static continuous phase, Egbuna [18] reported increases in mass-transfer coefficients of up to 80 per cent for drops in free fall, with an applied field strength of 3 kV/cm. Bailes [17] using both single and multi-drop extraction cells, showed that the overall mass-transfer coefficients, that is during both formation and free fall of the drops, were increased by 20–80 per cent for applied fields of up to 1 kV/cm. Although these significant enhancements of mass transfer have been obtained in liquid–liquid systems, the effect of electric fields on the mass transfer from droplets falling in a counter-current gas stream has not been reported and hence the present investigation has been undertaken.

EXPERIMENTAL

A test facility was constructed consisting of a square cross-sectional column fabricated from four sections of 1·27-cm thick "Tufnol" each 57 cm by 12·7 cm. Each side was fitted with a "Perspex" window 0·16-cm thick to enable observation and photography of the drops and the top and bottom of the column were sealed with square "Tufnol" plates and soft rubber gaskets. The upper electrode was fabricated from brass 0·64-cm thick and was connected to the high voltage generator by means of a sleeve arrangement which permitted movement giving electrode gaps infinitely variable up to 30 cm. The electrode was drilled with 8 0·64-cm dia. holes to facilitate passage of the gas phase and 1 0·79-cm hole permitting the brass nozzle to be positioned flush with the bottom of the electrode. Four nozzles were used, 0·107, 0·173, 0·235 and 0·313 cm in diameter. At the bottom of the column, a specially developed collecting device was used as the lower electrode. This consisted of an earthed circular copper vessel, 7·6-cm dia. through which water flowed, which was agitated by a stirrer driven from below in such a way as to produce a vortex into which the drops fell. Tests with drops containing a dye showed that on contact with the water surface, these were immediately dispersed and carried away with the water. The bottom plate of the column was drilled with two 0·64-cm holes to allow the gas phase to enter the column. For carrying out charge measurements, the drop collecting device was replaced by an earthed cathode consisting of a plate with a central copper wire gauze through which the drops fell. The gauze was connected via a d.c. amplifier to a recorder and this arrangement enabled charge in the range 0–10^{-7} C to be measured in increments of 10^{-11} C.

In the design of the test section, two configurations were considered. Firstly a point to plane design, in which the potential is applied to an isolated nozzle from which a drop falls through a field established by an earthed electrode below the nozzle. In such a system, the field would be strongly divergent and as the drop would follow the field lines, this would cause problems in collection. In addition, the field at the nozzle would be high, resulting in a limited range of potentials before drop disintegration took place. For these reasons, the second alternative, that of a plane to plane system, whereby the nozzle was surrounded by a guard electrode at the same potential, was adopted. Although the field between two parallel plates is not perfectly uniform due to end effects, the field at the vertical axis will be substantially uniform, leading to a simpler analysis of the system, providing the ratio of electrode width to separation is low. Taking this into account and the need to have a separation large enough to ensure a measurable droplet evaporation, the present geometry was evolved which is shown in Fig. 1.

Preliminary experiments showed that water formed satisfactory drops in the presence of fields, but the amount of evaporation was very low, whilst acetone provided measurable evaporation rates, though drop formation was erratic. Heating the water stream proved unsatisfactory and hence tests were carried out using 50 per cent w/w acetone–water mixtures as a compromise and nitrogen as the counter-current gas stream for reasons of safety. Nitrogen from a cylinder was passed through the column at rates of up to $35\,\mathrm{cm^3/s}$ and flows of liquid of up to $0{\cdot}02\,\mathrm{cm^3/s}$ were accurately metered under conditions of constant temperature. Column temperatures and pressures were noted and the d.c. power was provided using a "Brandenburg" generator (0–50 kV); the nozzle potential being measured with an electrostatic voltmeter. Still photographs of drops were taken using an electronic flash unit with a flash duration of 3 µs and cinephotographs were obtained at 400 frames/s. Development tests showed that accurate analysis of the liquid phase for acetone before and after passage through the column produced irreproducible data and a technique was developed in which the evaporated acetone was absorbed from the liquid phase in water. The acetone was then precipitated as dinitrophenyl hydrazone by a method based on that of Lappin and Clark [23], which was then evaluated spectrophotometrically. As finally adopted the method could be used to determine concentrations of acetone as low as $0{\cdot}003\,\mathrm{mg/cm^3}$ with an accuracy of ±2 per cent. In carrying out tests, care was taken to ensure the column was always swept out with air before starting a run and 0·5 ks was allowed for the attainment of steady state conditions.

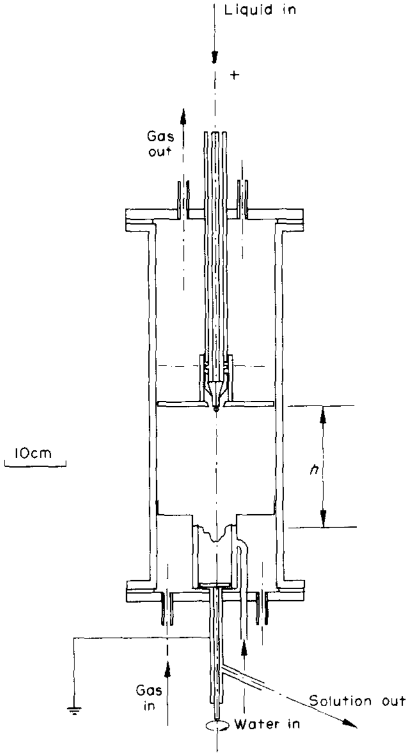

FIG. 1. Schematic diagram of test facility.

RESULTS

During measurements of mass transfer, it was noticed that a very small secondary droplet was formed from the disintegrating neck of the main drop. Close examination showed that at low field strengths, this droplet followed the main drop and was collected in the usual way. At high field strengths however, although the main drop was not affected, the now non-uniform field diverted the secondary droplet from its vertical fall and this eventually returned to the upper electrode. This was presumably due to evaporation reducing the gravitational force and the fact that the droplet was charged in the opposite sense to the main drop. Measurements were therefore limited to field strengths at which the secondary droplet was not deflected from the vertical axis.

Although the charge density induced on an ideal drop in an infinite parallel plate system is proportional to the applied potential gradient, the charge data obtained appeared to be a function of nozzle diameter and electrode spacing in addition to the applied gradient. It is considered that this was due to the compromise design of the test facility as discussed previously, resulting in uniform electric fields being produced only at electrode spacings of up to 5 cm. Even at this separation, charge densities were some 50 per cent below predicted values, probably due to maldistribution of charge across the surface of the drop, with, in the case of a drop formed at a nozzle, the greater part residing on the front or lower surface. In addition, it was observed that the drop at the nozzle became less spherical with increase in nozzle diameter and this is a significant factor contributing to the induced charge. Stewart [3] showed that the induced charge density increases with nozzle size with charged water droplets in *n*-heptane, which confirms the present observations. Obviously, as discussed by Thong *et al.* [19, 20], the charge induced on a drop has a vital role in determining the velocity in a field, though in view of the wide variations not only in gradient between the electrodes but also in drop shape, it was considered unrealistic to predict the induced charge and the velocity by the methods proposed by these works; but to use values obtained photographically as discussed later. In further work, it would obviously be of value to strive for a linear field distribution, though the problems of achieving measurable rates of evaporation must be a significant factor.

Considering hydrodynamic measurements, drop dimensions were obtained using a film analyser with a magnification of 500 × and measuring the major and minor axes. Typical drop volumes obtained in this way are shown in Fig. 2 and all the data are shown in Fig. 3 which shows the fractional decrease in the equivalent spherical drop diameter as a function of the potential gradient. This parameter calculated as:—

$$\Delta d_e = 1 - (d_e \text{ with field}/d_e \text{ at zero potential}) \qquad (2)$$

was found to be largely independent of nozzle diameter and Fig. 3 represents averaged data. The time taken for a drop to form at the nozzle was obtained directly for a known number of drops and the time for drop fall from analysis of the cine-film. Within the confines of the laboratory, it was not possible to erect a column in which terminal velocities were attained and it should be noted that the drops were accelerating throughout their flight. With no applied field, an average velocity was obtained from the area under the graph of instantaneous velocities and electrode spacing and the values given in Table 1, show that over the range of conditions considered, the average values are largely independent of nozzle diameter. For the case of applied fields, data were available at constant potential rather than field strength and the average velocity was calculated as:

$$u = u_0\,(u_a \text{ for applied field}/u_a \text{ at zero potential}) = u_0 \Delta u \text{ cm/s}. \qquad (3)$$

A good correlation of all the averaged data was given by:

$$\Delta u = 0{\cdot}074(V/h)^{1{\cdot}57}. \qquad (4)$$

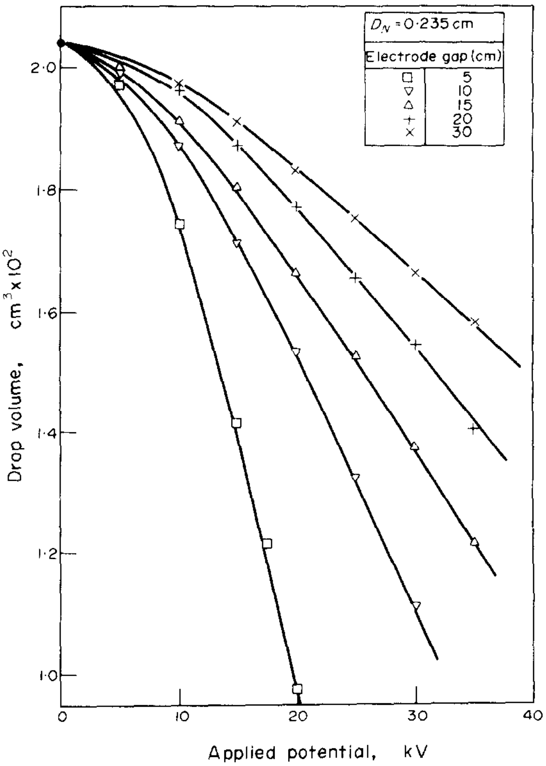

FIG. 2. Drop volumes as a function of applied potential at different electrode gaps.

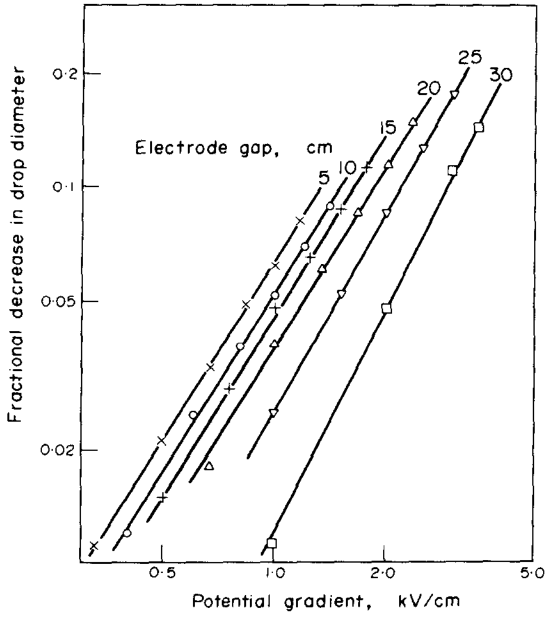

FIG. 3. Fractional decrease in equivalent spherical drop diameter, Δd_e, as a function of potential gradient.

Table 1. Average drop velocities at zero potential

Electrode gap (h cm)	Average drop velocity during flight (cm/s)				
	D_N (cm) 0·107	0·173	0·235	0·313	Mean value
5	48·4	48·8	48·8	48·4	48·6
10	82·4	82·8	82·4	81·3	82·2
15	108·1	108·5	108·9	107·2	108·2
20	136·1	131·5	131·6	129·7	131·6
25	155·0	154·0	154·0	148·6	152·9
30	173·6	173·9	174·8	168·8	172·5

Using the drop velocity data, values of $Re = d_e u\rho/\mu$ were calculated and for all the nozzles the data were correlated in a similar way to the mean velocities as shown in Fig. 4 as:

$$\Delta Re = 0{\cdot}032(V/h)^{1{\cdot}51}. \qquad (5)$$

The mass-transfer data were all expressed as $M = m/y_A$ and for the case of zero potential, a plot of M against electrode gap produced a linear relation with a finite intercept from which the percentage of evaporation taking place at the nozzle could be obtained. Using these percentages, it was possible to obtain the mass transferred during fall alone as distinct from the total mass transfer and express this in terms of a coefficient and hence a Sherwood number defined as:

$$Sh = (M/(M_w tA))(RT/P)d_e/D_v \qquad (6)$$

where D_v, the diffusivity of acetone in nitrogen was calculated as 0·1005 cm^2/s. A plot of Sh and Re showed that the data for charged drops coincide with those for uncharged drops suggesting that the most significant effect of electric fields was to increase the Reynolds' number and the wide scatter of data suggested that a further parameter should be taken into account. Working on this basis, it was found that the empirical relationship:

$$Sh = 3{\cdot}0 + (0{\cdot}345 d_{e_0} - 0{\cdot}0744)Re_0(1 + 0{\cdot}032(V/h)^{1{\cdot}51}) \qquad (7)$$

provided a good correlation of all the data, both for charged and uncharged drops as shown in Fig. 5.

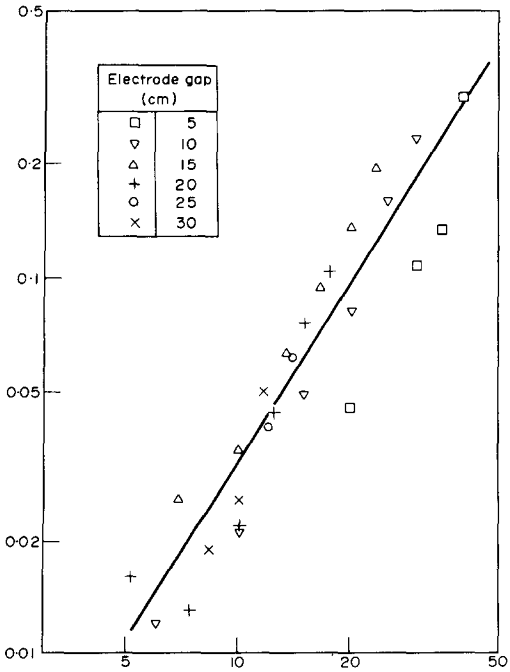

FIG. 4. Fractional increase in Reynolds' number, ΔRe, as a function of potential gradient.

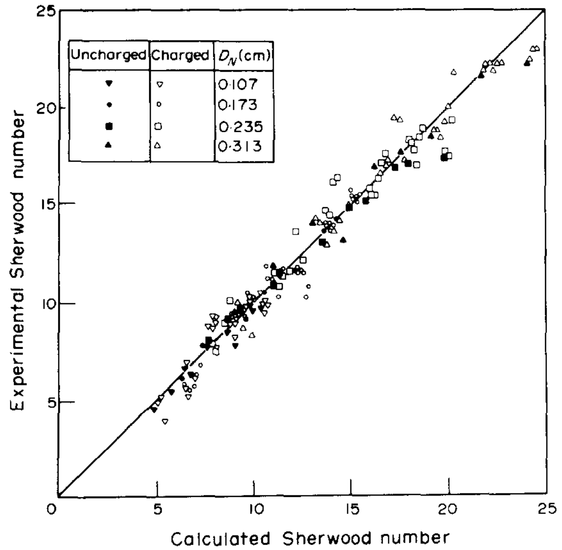

FIG. 5. Experimental values of Sherwood number compared with values calculated from the equation: $Sh = 3{\cdot}0 + (0{\cdot}345 d_{e_0} - 0{\cdot}0744)Re_0(1 + 0{\cdot}032(V/h)^{1{\cdot}51})$.

CONCLUSIONS

The main conclusions which may be drawn from this investigation are as follows:

(i) At constant electrode spacing, increase in the applied potential, decreases the drop size, increases the drop velocity when it has left the nozzle resulting in a net increase in Reynolds' number.

(ii) The field created by applying a high voltage at the upper electrode was linear for an electrode gap of 5 cm and became less uniform with increased electrode spacing.

(iii) The drop size decreased with increased electrode spacing due to non-linear fields creating a high field strength around the nozzle thus decreasing the effective interfacial tension of the drop. With a non-linear field, the drop velocity is a function of the applied potential gradient only.

(iv) The increase in drop velocity is relatively greater than the decrease in drop size resulting in an increase in Reynolds' number given by:

$$\Delta Re = 0{\cdot}032(V/h)^{1{\cdot}57}. \qquad (7)$$

(v) Mass-transfer rates from uncharged falling drops increase with drop size due to an increased oscillation effect. The data are well correlated by:

$$Sh = 3{\cdot}0 + (0{\cdot}345\, d_e - 0{\cdot}0744) Re. \qquad (8)$$

This may be compared with the frequently quoted expression of Ranz and Marshall [24] derived for drops suspended at a capilliary:

$$Sh = 2{\cdot}0 + 0{\cdot}6\, Re^{0{\cdot}5} Sc^{0{\cdot}33}. \qquad (9)$$

Mass-transfer rates for charged drops are increased by up to 50 per cent by applying a potential gradient of 3·5 kV/cm; the increase being almost entirely due to the increase in Reynolds' number with only very small enhancement of internal circulation and interfacial turbulence effects.

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EFFET DU CHAMP ELECTRIQUE SUR LE TRANSFERT MASSIQUE DE GOUTTES TOMBANTES

Résumé—Des études sur l'effet du champ électrique sur le transfert massique ont été limitées aux systèmes liquide–liquide. On mesure ici l'effet de gradients de potentiel, compris entre 0 et 3,5 kV/cm, sur le flux d'évaporation d'acétone à partir de gouttes d'une solution à 50 pour cent acétone–eau dans un contre-courant d'azote. On donne des précisions sur la taille des gouttes, la charge, la vitesse des gouttes, le temps de résidence et on conclut que l'accroissement du transfert massique, jusqu'à 50 pour cent, est du à l'accroissement du nombre de Reynolds plutôt qu'à l'augmentation de la turbulence interfaciale et à la circulation interne.

DER EINFLUSS ELEKTRISCHER FELDER AUF DEN STOFFTRANSPORT BEI FALLENDEN TROPFEN

Zusammenfassung—Untersuchungen des Einflusses elektrischer Felder auf den Stoffübergang haben sich bisher im wesentlichen auf flüssig/flüssig-Systeme beschränkt.

In dieser Arbeit ist nun der Einfluß von Feldstärken im Bereich von 0 bis 3,5 kV/cm auf die Verdampfung von Azeton aus Tropfen einer 50% Azeton-Wasser-Lösung im entgegenströmenden Stickstoff gemessen worden. Angaben über Tropfengröße, Beladungsdichte, Tropfengeschwindigkeit und Verweilzeit werden mitgeteilt. Aus den Ergebnissen wird der Schluß gezogen, daß die beobachteten Steigerungen des Stoffüberganges, die bis zu 50% betragen, eher auf größere Reynolds-Zahlen als auf eine Steigerung der Turbulenz an der Phasengrenze und interne Zirkulation zurückzuführen sind.

ВЛИЯНИЕ ЭЛЕКТРИЧЕСКИХ ПОЛЕЙ НА МАССОПЕРЕНОС ОТ ПАДАЮЩИХ КАПЕЛЬ

Аннотация — Исследовано воздействие электрических полей на массоперенос в опытах с системами жидкость-жидкость. Выявлено влияние градиентов электрического потенциала в диапазоне 0–3,5 кв/см на скорость испарения ацетона из капель 50% водного раствора ацетона во встречный поток азота. Приводятся данные по размерам капли, заряду времени их существования и делается вывод о том, что интенсификация массопереноса на 50% скорее объясняется возрастающими числами Рейнольдса, нежели усилением межфазной турбулентности и внутренней циркуляции.