

contrary, while substances of relatively low oil affinity but which penetrate readily into and disorder the lipid microcrystalline structure may display exceedingly high skin permeability. Such substances should be able to cause marked increases in D_L , not only for themselves, but also for other concurrently permeating species. Indeed, it seems probable that universal solvents such as dimethyl sulfoxide and hexamethyl phosphotriamide, which permeate through intact skin at phenomenally rapid rates and substantially enhance the permeation of other substances, may operate by creating controlled disorder in organized lipid membranes.

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NOTATIONS

a	= activity
C	= concentration
D	= diffusion coefficient
J	= flux
k	= partition coefficient
K	= ionization constant
l	= length
P	= permeability
R	= resistance
t	= thickness
x	= distance

Greek Letters

Δ	= difference operator
τ	= thickness of plate element
δ	= thickness of interstitial layer
α	= l/τ = plate axial ratio
β	= δ/τ
ϕ	= volume fraction
σ	= lipid phase/protein phase partition coefficient

Subscripts

Aq	= aqueous phase
B	= unionized base form
BH ⁺	= ionized form
H ⁺	= hydrogen ion
L	= lipid phase
M	= membrane
max	= maximum

P	= protein phase
S	= stratum corneum

LITERATURE CITED

- Anderson, R. L., and J. M. Cassidy, "Variations in Physical Dimensions and Chemical Composition of Human Stratum Corneum," *J. Invest. Dermatol.*, **61**, 30 (1973).
- Blank, I. H., "Penetration of Low-Molecular Weight Alcohols into Skin. I. The Effect of Concentration of Alcohol and Type of Vehicle," *ibid.*, **43**, 415 (1964).
- , R. J. Scheuplein, and D. J. MacFarlane, "Mechanism of Percutaneous Absorption. III. The Effect of Temperature on the Transport of non-Electrolytes across the Skin," *ibid.*, **49**, 582 (1967).
- Holbrook, K. A., and G. F. Odland, "Regional Differences in the Thickness of the Human Stratum Corneum: An Unstructured Analysis," *ibid.*, **62**, 415 (1974).
- Hunter, J. A. A., "Diseases of the Skin: Structure and Function of Skin in Relation to Therapy," *Brit. Med. J.*, **4**, 340 (1973).
- Katz, M., and B. J. Poulson, "Absorption of Drugs through the Skin," in *Handbook der Experimentellen Pharmacologie; Concepts in Biochemical Pharmacology*, Part I, B. B. Brodie and J. R. Gillett, ed., Springer Verlag, New York (1971).
- Mackenzie, I. C., and J. E. Linder, "An Examination of Cellular Organization within the Stratum Corneum by a Silver Staining Method," *J. Invest. Dermatol.*, **61**, 245 (1973).
- Montagna, W., and P. F. Parakkal, *The Structure and Function of Skin*, 3 ed., Academic Press, New York (1974).
- Rein, H., "Experimental Electroendosmotic Studies on Living Human Skin," *Z. Biol.*, **81**, 125 (1924).
- Rothman, S., *Physiology and Biochemistry of the Skin*, The University of Chicago Press, Ill. (1965).
- Scheuplein, R. J., "Mechanism of Percutaneous Absorption. I. Routes of Penetration and the Influence of Solubility," *J. Invest. Dermatol.*, **45**, 334 (1965).
- , "Mechanism of Percutaneous Absorption. II. Transient Diffusion and the Relative Importance of Various Routes of Skin Penetration," *ibid.*, **48**, 79 (1967).
- , "Molecular Structure and Diffusional Processes across Intact Epidermis," Edgewood Laboratory, Contract Report 18 (1967).
- , and I. H. Blank, "Permeability of the Skin," *Physiol. Rev.*, **51**, 702 (1971).
- , "Mechanism of Percutaneous Absorption. IV. Penetration of non-Electrolytes (Alcohols) from Aqueous Solutions and from Pure Liquids," *J. Invest. Dermatol.*, **60**, 286 (1973).
- Wurster, D. E., "Activation Energy Required for Skin Penetration," Final Contract Report to U.S. Army Chem., R&D Laboratories, Contract DA18-108-AMC-168 (1964).

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Breakup of a Turbulent Liquid Jet in a Low-Pressure Atmosphere

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Breakup length measurements were made of turbulent liquid jets in a low-pressure atmosphere. As a result of these experiments, it was found that the jets obey a pseudo laminar analogy only at low exit velocity. The nondimensional breakup length correlates with the jet Weber number through the complete range.

The breakup process of turbulent liquid jets has received little attention in the scientific literature. In a previous paper concerning this topic, the author, Phinney (1973), recounted some of the history and difficulties connected with the investigation of this problem. An analogy was postulated to exist between the breakup of laminar and turbulent liquid jets. A conclusion from this analogy

is that the ambient atmosphere should have an influence on the breakup of the jet. Because of the absence of accurate data concerning breakup in a low-density environment, this conclusion could not be adequately tested. A recent set of experiments was conducted to determine the threshold below which the ambient density becomes unimportant. These experiments are the turbulent equivalent of those of Fenn and Middleman (1969) for laminar jets.

CONCLUSIONS AND SIGNIFICANCE

The jet breakup length was found to be independent of the ambient gas density. The new data, together with those from other sources, show that nondimensional breakup length (L/D) is a function of the jet Weber number We_j only. The low-velocity data relate (L/D) linearly with We_j , corresponding to the proposed pseudo laminar analogy. The remaining data, the high velocity portion, are correlated by the same parameter We_j but along a

different line segment corresponding to a truly turbulent regime. A simple analysis shows that both regimes should indeed be governed by the same parameter and that the ambient pressure should have no influence through the range of these experiments as was observed. Besides its practical importance for design, the data correlations are useful because they help identify the existence of two regimes of breakup, the boundary between them, and the physical mechanism that controls each mode of breakup.

EXPERIMENTAL APPARATUS AND PROCEDURE

The present experiments are a simple extension of those reported in a previous paper by the author (1973). The electrical gate circuit that detects breakup is identically the same piece of apparatus as was used before, as is the pressure system that feeds the jet. What was changed is that the jet and detection screen are surrounded by a Plexiglas cylinder with aluminum end plates in order to form a vacuum chamber. The inside diameter of the cylinder is 28 cm; the length is 120 cm. The pressure can be reduced to the order of 6.5 kN/m² with a small vacuum pump.

A schematic diagram of the vacuum system is shown in Figure 1. Not shown, in order to simplify the drawing is a recharging system of valves and pipes that permit the sump to be cut off and pressurized in order to force the fluid through a pipe back into the jet supply tank. Also not shown are the electrical components which consists of a gate circuit that is controlled by conduction of a small current through the jet. The percentage of the time that the jet is broken can be read directly on a counter which is connected to a pulse generator through the gate circuit. The gate circuit makes the measurement independent of the electrical resistance of the jet. The screen that detects the breakup point of the jet is moved by a lead screw that is brought through the end plate by a rotary seal. The position of the screen is read directly from a scale in the vacuum chamber. A tare length to be subtracted from subsequent length measurements is obtained by bringing the screen into contact with the end of the jet nozzle and by reading the scale. The accuracy of the length measurements is about 1 mm.

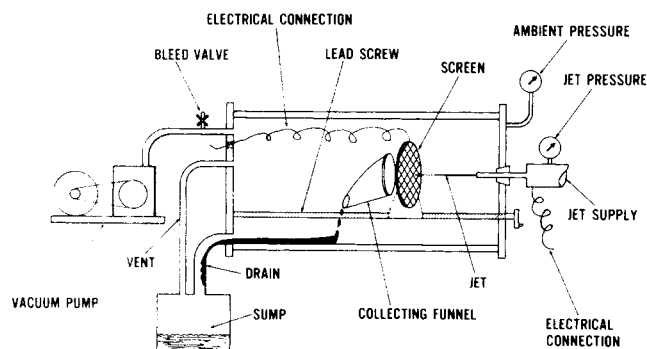


Fig. 1. Schematic diagram of experimental apparatus.

TABLE 1

Nozzles	Diameter (cm)	Length (cm)
B	0.206	20.3
G	0.1024	10.5

Two cylindrical nozzles were made from glass capillary tubing. Their dimensions are given in Table 1. Nozzle B is the same as that used in the previous paper.

The fluid is a solution of table salt with properties as follows: the jet density $\rho_j = 1.062$ g/cm³, the jet viscosity $\mu = 1.15$ centipoise, and the surface tension $\sigma = 73.1$ dyne/cm. The density is determined from weight volume measurements, the viscosity from flow measurements through a long capillary tube, and the surface tension by means of a Du Nöuy tensiometer.

For given flow conditions, the screen was moved until a position was found where the pulse counter showed that the jet was broken 50% of the time. This was taken to be the breakup distance for that condition. For each such datum point, the breakup length, the ambient pressure, and the jet supply pressure were recorded. From a previously performed calibration, the jet exit velocity can be determined from the nozzle pressure difference.

EXPERIMENTAL RESULTS

By using the test procedure outlined above, data were obtained from the two nozzles described in Table 1. The atmosphere was air, and a series of tests were run with the ambient pressure set at 93.3, 33.3, and 9.3 kN/m². The raw data were reduced to the following dimensionless parameters: nondimensional breakup length (L/D), the jet Weber number We_j , the ambient Weber number We_a , and the Reynolds number Re . An examination of the effect of ambient density on the breakup length did not show the expected correlation with ambient Weber number We_a (there was, in fact, no influence of ambient density at all). Figure 3 of the previous paper, Phinney (1973), shows the lack of correlation in terms of Re .

Eliminating We_a and Re as correlating parameters for (L/D), we are left with We_j . The data are plotted in terms of these nondimensional parameters in Figure 2

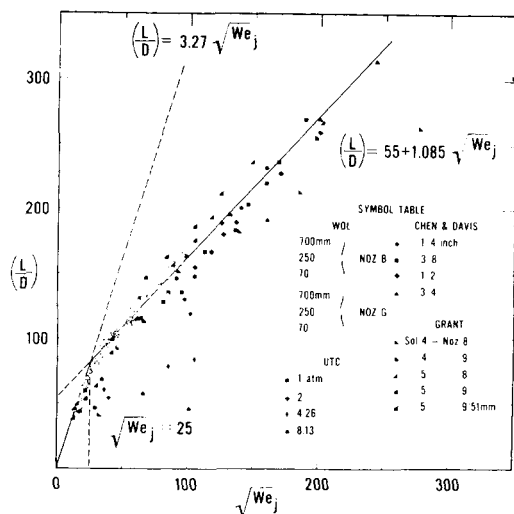


Fig. 2. Correlation of experimental data.

(alternate points have been omitted near the knee of the curve to avoid cluttering the figure). This presentation is also similar to that of Figure 12 of Grant and Middleman (1966), except that Cartesian coordinates are used instead of a log-log plot. The present method of displaying the data in a linear plot emphasizes the low-speed portion in such a way that a break in the curve is disclosed which might not be apparent otherwise.

Two line segments are drawn in Figure 2 which are fitted to the data by a least-squares procedure. The low-velocity segment is forced to go through the origin. Its slope is determined by the nine data points with $\sqrt{We_j} \leq 20$. The high-velocity portion is determined by the 105 data points with $\sqrt{We_j} \geq 35$. The mean-square error (in L/D) for the low-velocity correlation is 2.3, and it is 4.4 for the high-velocity segment. The intersection of the two lines gives a critical Weber number $\sqrt{We_{jcrit}} = 25$ for the boundary between pseudo laminar and "truly turbulent" behavior.

Data of Chen and Davis (1964) and the unpublished data from UTC are also included in Figure 2. Some additional data of Grant and Middleman (1966) are likewise included. The data does not seem to show any systematic departure from the line segments that are drawn. The data from the various sources are fairly consistent. The White Oak Laboratory data show very good correlation, independently of the ambient pressure. The UTC data that were used to establish the ambient density effect in the previous paper, Phinney (1973), now appear scattered, but not in any systematic way with ambient density.

ANALYSIS OF RESULTS

An examination of the present data reveals an important feature that was not considered in the previous paper, Phinney (1973). Unlike laminar jets, it is necessary to introduce a parameter to describe the magnitude of the internal random motion of the jet. The following argument demonstrates the need for such a parameter. The internal motion can be sufficiently violent that the dynamic pressure associated with it becomes greater than the pressure due to surface tension. At this point, the jet will rupture because of the surface tension's inability to contain the motion. As will be shown shortly, the velocity of the turbulent internal motion increases proportionally to the jet exit velocity, while the surface tension force remains constant. As a result, there must be some threshold velocity above which breakup is caused by the internal turbulent motion. It is interesting to note that surface tension

and the internal motion interchange roles as the above-mentioned threshold is crossed. In a laminar (or pseudo laminar) jet, the contraction of surface tension is the disruptive force, and the internal motion is the stabilizing force in that it provides the inertia that delays breakup. On the other hand, if the internal motion is great enough, then the surface tension acts as a retarding force which tends to smooth a roughened surface, while it is the turbulent motion which produces protuberances that tend to disrupt the jet.

Let v_i stand for the root-mean-square value of a velocity that is typical of the turbulent motion in the jet. A Weber number based on v_i , $\rho_j v_i^2 D / \sigma$, gives the ratio of the dynamic pressure of the turbulent motion to the pressure connected with the surface tension. The magnitude of v_i can be estimated by the fact that in pipe flow (which represents the nozzle flow before and hopefully after the exit) the turbulence intensity $I = v_i / V$ is found experimentally to be a very mild function of the Reynolds number Re . If one assumes for the moment that the turbulence intensity I is constant, then v_i is proportional to V , and hence, the square roots of the Weber numbers based on v_i and V are also proportional. By this reasoning, it follows that the truly turbulent and the pseudo laminar cases both correlate on the basis of the same parameter We_j . This observation agrees with the fact that the presentation of Figure 2 appears to correlate the data. The threshold for the truly turbulent regime would seem to be at $\sqrt{We_j} = 25$, where the break between the two segments is found in Figure 2. Furthermore, below the threshold, the breakup length increases linearly with $\sqrt{We_j}$, as predicted by laminar theory. In this pseudo laminar regime, the slope of the line is 3.27, whereas for a truly laminar jet, the slope is found to be in the range of 10 to 15 by various authors. Laminar theory identifies the slope as the logarithm of the inverse of the initial disturbance level. The lower value for the pseudo laminar case indicates a higher initial disturbance level.

A constant turbulence intensity implies that the actual turbulent velocity and the corresponding fluctuating dynamic pressure both increase with the discharge velocity. At low exit velocity, we have the picture of a turbulent flow in which the dynamic pressure is negligible. This is the pseudo laminar case in which there is a constant input disturbance level, and the long wavelength surface roughness is amplified by the pinching off action of the surface tension. As the velocity increases, a threshold is reached where the fluctuating dynamic pressure of the turbulence is of the same order as that due to the surface tension. Above the threshold, the turbulent motion becomes dominant and changes the mechanism of breakup.

The physical interpretation given above is consistent with the observation that the experimental data in Figure 2 do not show any apparent influence of the ambient pressure. Assume that for data in the pseudo laminar regime, the influence of the ambient density is the same as it is for strictly laminar jets. If this is so, then according to Fenn and Middleman (1969), the critical Weber number based on ambient density We_a should be 5.3. Since none of the data in this regime ($We_j < 25$) had We_a that large, no ambient influence should have been expected. For the truly turbulent regime, there is a point at which the ambient density becomes large enough that the dynamic pressure connected with the relative motion between the jet and the atmosphere becomes of equal magnitude with the dynamic pressure of the internal motion. This critical point would occur for an ambient density such that $\rho_a V^2 > \rho_j v_i^2$. Turbulence measurements in a pipe suggest the turbulence intensity $I = v_i / V$ would be

on the order of 0.05, which gives a value of (ρ_a/ρ_j) below that for the experiments in Figure 2. On this basis there should be no observed effect in the truly turbulent case either.

CONCLUDING REMARKS

The experiments herein reported show that the analysis given in the preceding paper, Phinney (1973), was incomplete. In Figure 5 of that paper, what was purported to be the influence of the ambient Weber number We_a on breakup length should be interpreted as the variation due to the jet Weber number We_j . The failure to be able to distinguish between the two parameters was due to the fact that almost all of the data were taken with the jet fluid being water and the ambient gas being atmospheric air. The new experiments suggest the necessity of an additional breakup mechanism that was not considered in the previous paper. The new parameter can be thought of as representing a new mode of breakup, although it reduces to We_j which governs pseudo laminar breakup (with no ambient influence) as well. A large portion of the existing experimental data is in the range of this new mode of breakup. The pseudo laminar breakup that was described in the previous paper does appear to be a valid picture of the breakup of turbulent jets with low enough exit velocity, $\sqrt{We_j} < 25$. The influence of the ambient atmosphere was not observed, although it should be present in either mode of breakup if the ambient density is high enough.

The remarks in the previous paper, Phinney (1973), concerning the optical observations of breakup still stand and are consistent with the present work. Near the exit, the turbulent motion produces protuberances which give the surface a rough spiky appearance. Since the surface tension force normal to the surface is inversely related to the radius of curvature, the sharper the protuberance the more strongly it is contained. For this reason, it is likely that at low velocity, where the turbulent motion is only somewhat above the threshold, the ultimate breakup of the jet will be due to fairly long wavelength disturbances just as in the case of pseudo laminar breakup. General observations show that as the exit velocity is increased (and the turbulent intensity becomes higher), the breakup becomes more violent and the scale and droplet size decrease.

The statistical measurements that were presented in Figure 6 of the previous paper span the range from the

pseudo laminar to the truly turbulent. In particular, the low Reynolds number runs for cases *CI* and *DI* are just below the break in the curves; case *BI* ($Re = 8500$) is at the break, and all the rest are at some point above the break. All these data seem to correlate in terms of the parameters in that figure.

Although the present data did not show an ambient density influence, there must be a threshold above which this effect exists and could be studied. It would be useful to perform these additional experiments. Unfortunately, because of the design of the apparatus, it was not possible to pressurize it, thus making the higher range of We_a inaccessible.

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NOTATION

D	= nozzle diameter
I	= turbulence intensity, v_i/V
L	= jet breakup length
Re	= jet Reynolds number, $\rho_j V D/\mu$
v_i	= root-mean-square value of a velocity typical of the jet internal motion
V	= mean jet exit velocity
We_a	= ambient Weber number, $\rho_a V^2 D/\sigma$
We_j	= jet Weber number, $\rho_j V^2 D/\sigma$
μ	= viscosity coefficient of jet fluid
ρ_a	= density of ambient gas
ρ_j	= density of jet fluid
σ	= surface tension

LITERATURE CITED

- Chen, T. F., and J. R. Davis, "Disintegration of a Turbulent Water Jet," *Proc. Am. Soc. Civil Eng.*, **HY1**, 175 (1964).
 Fenn, R. W., and S. Middleman, "Newtonian Jet Stability: The Role of Air Resistance," *AIChE J.*, **15**, 379 (1969).
 Grant, R. P., and S. Middleman, "Newtonian Jet Stability," *AIChE J.*, **12**, 669 (1966).
 Phinney, R. E., "The Breakup of a Turbulent Jet in a Turbulent Jet in a Gaseous Atmosphere," *JFM*, **60**, 689 (1973).

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Transient Viscoelastic Flow of Polymer Solutions

Stress growth and stress decay data for two highly viscoelastic polymer solutions indicate that the first and second normal stress differences grow and decay at the same relative rates. These shear stress and first normal stress difference data are not represented well by the Spriggs, Bogue-Chen, or Carreau constitutive equations. The second normal stress difference, stress growth, and decay data are the first to be reported. The data were derived from radial pressure distributions and total normal and torsional forces obtained in cone and plate shearing. Sensitive miniature pressure transducers with pressure sensing diaphragms mounted flush with the plate surface provided the radial pressure distribution data.

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