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Electroconvection cells in dielectric liquids interfaced with conducting fluids

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[Plates 21–23]

Electroconvection cells at fluid/fluid interfaces may be part of a general pattern which includes convection cells driven by thermal energy transport and those driven by chemical potential transport. Interfacial tension appears to be the unstable parameter in each case.

Electroconvection cells were generated in thin layers of eight different isotropic dielectric liquids by bombarding the liquid/air interface with negative corona. The same electroconvection cells formed and followed the same sequence of development in each liquid. Three types of electroconvection cells were found, each being a ring vortex of liquid but with differing sizes and rotations.

Increasing current density was accompanied by decreasing diameters of types II and III cells, and by a gradual large increase in total surface area resulting from the nature and behaviour of type II cells which form open channels penetrating entirely through the liquid layer.

The vertex angle for II cell cones frozen in a thermoplastic recording compared favourably with a limiting value suggested by Taylor (1964) who may have measured II cell vertices in experiments which obscured cellular motion.

Introduction

While electroconvection cells have been known for nearly a century as scientific curiosities, they recently have assumed some practical importance. Electroconvection cells either participate directly in the function of, or establish performance limits for, a number of information oriented phenomena. Eidophor (Bauman 1953), thermoplastic recording (Glenn 1959), frost xerography (Gundlach & Claus 1963), liquid crystal displays (Penz 1970), and photoplastic recording (Aftergut, Kopczewski & Burgess 1971), are information display or recording processes which depend in some manner on electroconvection cells. Accordingly, both the detailed cell morphology, as well as the conditions required for the onset of electroconvection, are increasingly important.

Electroconvective phenomena are strongly influenced by the boundary conditions in a particular system. Boundary conditions can be divided into classes depending on whether the sign of the electric field is constant or alternating, on whether the dielectric liquid is isotropic or anisotropic, and on whether the system contains a single fluid (bounded by two semi-infinite solid electrodes) or contains the interface between two immiscible fluids with differing electrical properties (a dielectric fluid bounded by one solid electrode and the interface with a conducting fluid). All of the processes listed above, except liquid crystal displays, fall in the class of isotropic dielectric liquids bounded by one solid electrode and the interface with a conducting fluid having an electric field of constant sign between them. The recent studies of this class, e.g. Malkus & Veronis (1961), Watson, Schneider & Till (1970), and Atten & Moreau (1970) have all focused primarily on the conditions required for the onset of electroconvection and have given no attention to the details of cell structure or behaviour. The present report describes observations of cell morphology for this same class of boundary conditions.

The only previous description of cell morphology is that of Avsec & Luntz (1937) (A. &. L) who observed two different cell types in 2 mm thick layers of a dielectric liquid interfaced with air and exposed to d.c. corona. Both are ring vortices of liquid. The A. & L. 'a' cell has a concave centre at the interface and marked hexagonal boundaries. The A. & L. 'b' cell has a convex centre at the interface and marked hexagonal boundaries. The 'b' cell is confirmed by this study but not the 'a' cell. Significant new phenomena are reported here made possible by experimental conditions which produce cell boundaries stationary enough to permit leisurely observation.

It is important to note that electroconvection cells at fluid interfaces may be part of a more general pattern. Convection cells in liquids may also be propelled by thermal gradients according to Bénard (1900), as well as by chemical potential according to Orell & Westwater (1961) and Brian, Vivian & May (1971). A generalized summary of cellular convection phenomena might be: 'When a free boundary between two immiscible fluids must transmit energy in excess of a characteristic flux, convection cells may form to assist the transport.' The more interesting phenomena occur when energy is transported in a direction normal to the plane of the fluid interface.

For thermal energy transport across liquid layers less than 1 cm thick, Pearson (1958) has demonstrated that interfacial tension is the system parameter which becomes unstable and initiates cellular motion. Studies by Berg & Acrivos (1965) have supported and extended this conclusion. For chemical potential energy transport, Sternling & Scriven (1959) have demonstrated that interfacial tension is again the system parameter which becomes unstable. By analogy then one may expect interfacial tension to be the unstable parameter when a thin layer of dielectric liquid is spread on a conducting solid surface in air or vacuum and subjected to homopolar charge injection by corona or electron bombardment. Cressman (1963) has suggested that mutual repulsion forces between charges located at or near the liquid surface will act in opposition to interfacial tension forces. If this is the case, as the charge flux increases to the point where repulsion forces counterbalance interfacial tension forces, a large increase in liquid surface area may be expected. This increase in surface area is observed in the experiments reported here.

With mutual repulsion forces between charges of the same magnitude as interfacial tension forces, the perturbation of charge on a uniformly charged surface is equivalent to perturbing the net effective interfacial tension, and may similarly initiate cellular convection.

Experimental procedure

The equipment used consisted of a dissecting microscope and a variable high voltage power supply. A loop of 5 mil (0.13 mm) tungsten wire connected to the high voltage power supply was mounted below the microscope stage after removing the mirror. The glass microscope stage was used as the working surface after the bottom side was coated with a transparent conducting layer of tin oxide. In the centre of the stage a rectangular working area was defined by sandblasting the perimeter in such a way as to remove the tin oxide completely from a strip about 1 mm wide. The working area was therefore isolated electrically from the remainder of the stage. A hole drilled through the stage in one corner of the working area was filled with silver paste and a wire embedded from the top side making an electrical connexion to the working area without bridging the insulating boundary. The metal microscope frame was grounded as was the tin oxide coating outside the working area.

Dielectric liquids were spread on the working area in thicknesses from 3 to 30 μm then bombarded by uniform negative corona discharge from the tungsten wire loop. The insulating strip at the perimeter of the working area formed a charged boundary which dielectric liquids would not cross during bombardment. This feature prevents a rapid decrease in liquid layer thickness which otherwise occurs due to the pressure effect of charge on the liquid surface. The capability for creating steady-state conditions with stationary cell boundaries depends on maintaining a constant liquid thickness because both cell size and morphology vary with liquid thickness.

Achieving stationary cell boundaries depends as well upon having a uniform distribution of charge falling on the working area. It is conveniently adjusted by changing loop size and spacing while observing those electroconvection cells which will be referred to later as type III cells. Under uniform charge distribution and delivery rate type III cells are stationary regular hexagons of uniform size. A non-uniform charge distribution produces type III cells which are irregular polygons having from four to nine sides per cell and which co-exist in mixed populations. Also the frequent collapse of old type III cells and creation of new ones is observed.

Experiments were conducted with silicone oil, mineral oil, Apiezon oil B, 10C transformer oil, pyranol, Eidophor fluid, Quaker State motor oil, and a mixed aromatic oil. These various liquids all had viscosities in the range 0.5 to 5 Pa s and resistivities greater than $10^9\,\Omega$ cm.

Results

The same three types of electroconvection cells were found in each oil.

Type I cells are convex in cross-section with unmarked boundaries. Each cell is an individual ring vortex of liquid. Along the principal axis of the ring liquid moves toward the fluid interface with enough momentum to create a convex surface contour over the cell. The toroidal axis of the ring is stationary relative to the principal axis. Cell diameter is always equal to twice the liquid layer thickness.

Type II cells are conical in cross-section with unmarked boundaries. Each cell is an individual ring vortex of liquid. Along the principal axis of the ring liquid moves away from the fluid interface with enough momentum to create a concave surface contour over the cell. The toroidal axis of the ring rotates around the principal axis to create a spiralling ring vortex. Cell diameter is a function of liquid layer thickness and current density. With increasing current density cell diameter first increases to a maximum, then decreases. Maximum cell diameter is approximately 10 times liquid layer thickness.

Type III cells are convex in cross-section with marked boundaries. Each cell is an individual ring vortex of liquid. Along the principal axis of the ring, liquid motion is toward the fluid interface with enough momentum to create a convex surface contour over the cell. The toroidal axis of the ring is stationary relative to the principal axis. Cell diameter is maximum at the current density required for formation and decreases with increasing current density. Maximum cell diameter is approximately 30 times liquid layer thickness. Liquid velocities of approximately 0.5 mm/s were estimated in one type III cell by dark field observation of a tiny solid particle entrained in the cell. At high current density the high III cell population density exerts sufficient pressure on the cell boundary to convert its initially circular outline into a regular hexagon.

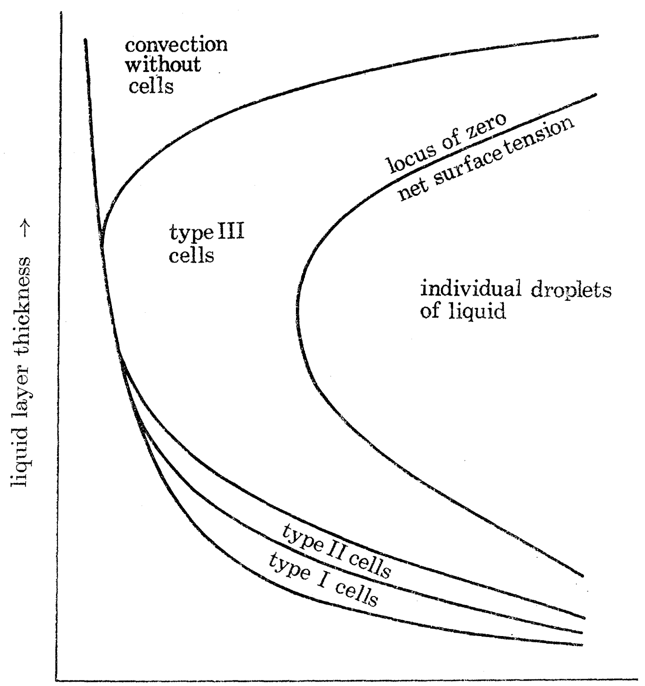


FIGURE 1. Qualitative relations for the three types of electroconvection cells.

An identical pattern of electroconvection cell development involving a sequential progression from one cell type to another in ascending order was observed in all liquids. In an approximate qualitative manner figure 1 illustrates the relations between liquid-layer thickness, current density, and cell type. Specific values of ordinate and abscissa depend heavily on liquid viscosity and resistivity and were not included in this exploratory study. The I cell and II cell regions are broad for the low viscosity, low resistivity liquids but were narrow and harder to detect in the others. Figure 1 also suggests a sharp boundary between I and II cells where in fact a substantial overlapping sometimes occurs.

As figure 1 indicates, thick liquid layers can develop type III cells only. At a lesser layer thicknesses II cells may be the first to develop followed by III cells at increased current density.

At liquid thicknesses generally between 3 and 30 μm cell types I, II and III occur sequentially and progressively with increasing current density. At the onset of convection, cell type I alone occurs. A further increase in current density will create a few II cell cones as shown in figure 2, plate 21. Occasionally a I cell will slide into a II cell cone and disappear on a downward spiral path. To the careful and patient observer transient behaviour is evident. The cell II cones gradually broaden and deepen to penetrate about half the fluid thickness, then emit a brief flash of light from the apex, followed by a decrease in size either to disappear or to repeat the sequence. By superimposing a pulse of current on the steady state value, one can cause many II cells to flash in unison about 0.1 s later as in figure 3, plate 21. This phenomenon will be referred to as 'II cell discharge'.

Further increase in current density causes the population of II cells to increase and neighbouring II cells to associate in forming random linear strings of cells defining ridges of liquid between adjacent strings. These ridges which are gnarled in appearance and occasionally bifurcated can be seen in figure 3 if one ignores the light flashes caused by the 'II cell discharge' which now occurs more frequently. Further current density increase again increases II cell population. Strings of II cells close ends to form circles as in figure 4, plate 22. 'II cell discharge' has become continuous. Each circle of II cells forms the boundary for a new III cell. Further increase in current density increases the population of III cells until population pressure straightens boundaries into regular hexagons (see figure 5, plate 22). The bright spots at III cell boundaries are remnants of II cells discharging continuously. The number of 'II cell discharges' bounding a III cell increases with increase in current density until they constitute almost the entire boundary between III cells. Still further increase in current density causes the III cell pattern to break up into individual drops of liquid, one per cell, each drop appearing to be spherical and in rapid random motion.

Discussion

The phenomenon called 'II cell discharge' is believed to actually be a discharge of charges collected near the surface of the liquid. It is thought not to be in the form of an arc or spark because the expected ultraviolet content is not present in the light flash, nor is the expected heat release found as the discharge becomes continuous, nor is broad-band radio frequency energy radiated. It seems much more likely and plausible that the flash of light results from the momentary creation of a geometrical discontinuity in the optical path such that a great deal of incident light is scattered.

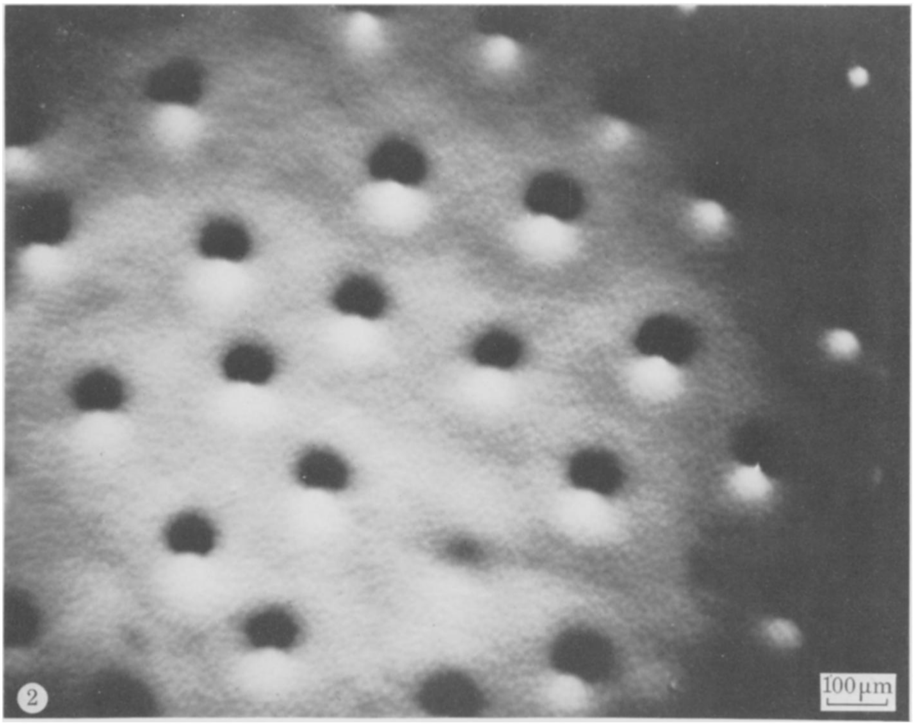
Remembering that in the final stage all of the liquid is divided into tiny individual droplets, it is evident that a large amount of new liquid surface area has been created. At the point where III cells convert to individual droplets only a differential increase in current density is required to cause the conversion. This is consistent with a gradual increase in surface area rather than with a sudden increase.

One can also suggest that the surface potential of the liquid stops increasing with further increases in current density once convection cells form in the liquid. C. W. Reed (personal communication) and Watson *et al.* (1970) confirmed this condition in systems where electrons traversed a vacuum to land on the surfaces of dielectric liquids to generate electroconvection cells. Consequently new conduction facilities are brought into use in direct proportion to further increases in current density.

Since new surface and new conduction facilities are created simultaneously by an increase in current density it seems probable that the former is responsible for the latter.

Gradual formation of new liquid surface area can occur through the following sequence of events. It begins with the transient formation of a tiny hollow channel from the apex of a II cell cone entirely through the remaining thickness of liquid to the tin oxide surface, the phenomenon previously referred to as 'II cell discharge'. Then as strings of II cells join ends to form circles bounding new III cells (figure 4, plate 22) these hollow channels lose their transient nature and become permanent. As III cells grow the channels grow broad and flat. Each bright spot on the III cell boundary in figure 5, plate 22 is a flattened channel extending from the free surface to the conducting base. III cells are isolated from their neighbours to the same degree that their boundaries are occupied by channels. In figure 5 one can see that the degree of isolation is extensive but not complete. After further increase in current density, the channels occupy the entire boundary around each III cell, isolation from neighbouring cells becomes complete, the cell assumes a spherical shape and moves rapidly across the conducting surface in contrast to the stable patterns of figure 5.

The idea that a II cell consists of a cone-shaped depression plus a narrow channel penetrating the remaining thickness of liquid is supported by the work of Taylor (1964) and Taylor & McEwan (1965). They found that the horizontal interface between a conducting fluid and a dielectric fluid becomes unstable as the strength of a vertical electrical field increases. A right circular cone of conducting fluid protrudes into the dielectric fluid, becomes increasingly acute, and after it attains a limiting vertex angle of 98.6°, discharges a microjet of conducting fluid from the vertex through the dielectric in the direction of the opposing electrode.



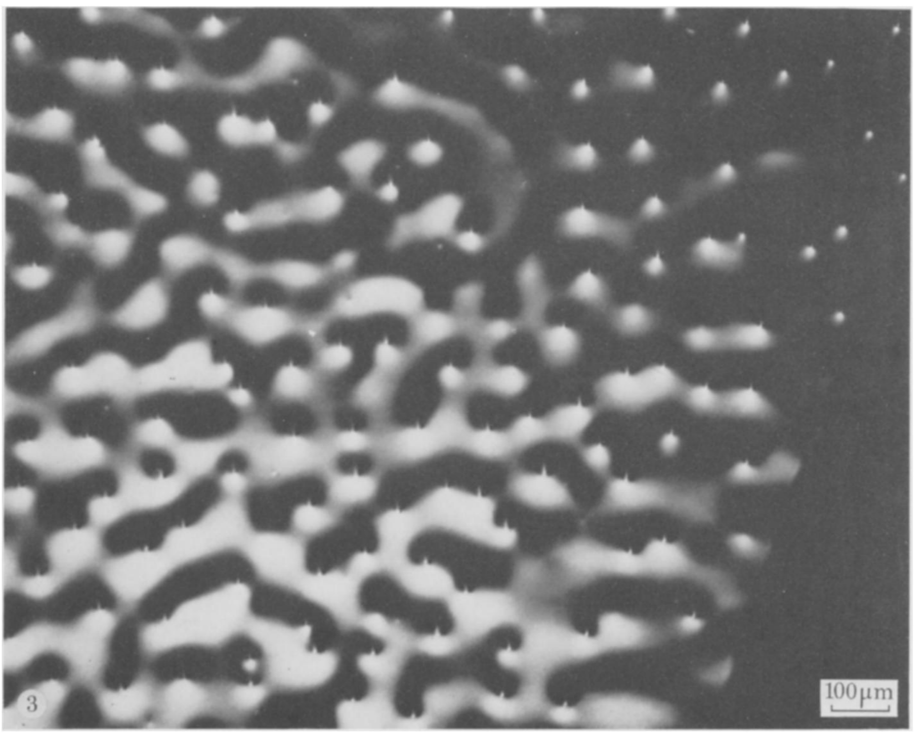


FIGURE 2. Large conical type II electroconvection cells against a background of small pebbly looking type I cells. Quaker State motor oil S.A.E. 30, 13 μm thick, 23 °C, 13 μA/cm^2.

FIGURE 3. Strings of type II electroconvection cells defining ridges of liquid. Dark areas are ridges and light areas are valleys. A current pulse applied milliseconds before this photograph has caused many of the II cells to flash or discharge simultaneously. Mixed aromatic oil, 5.5 μm thick, $10^{-3}\,m^2\,s^{-1}$, 33 °C, 129 μA/cm^2.

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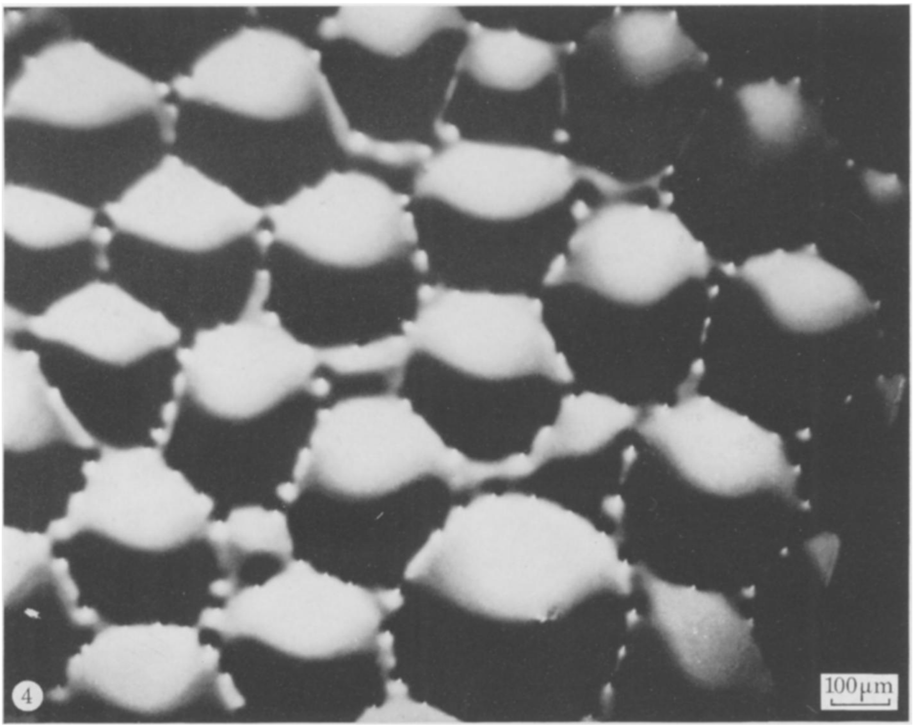

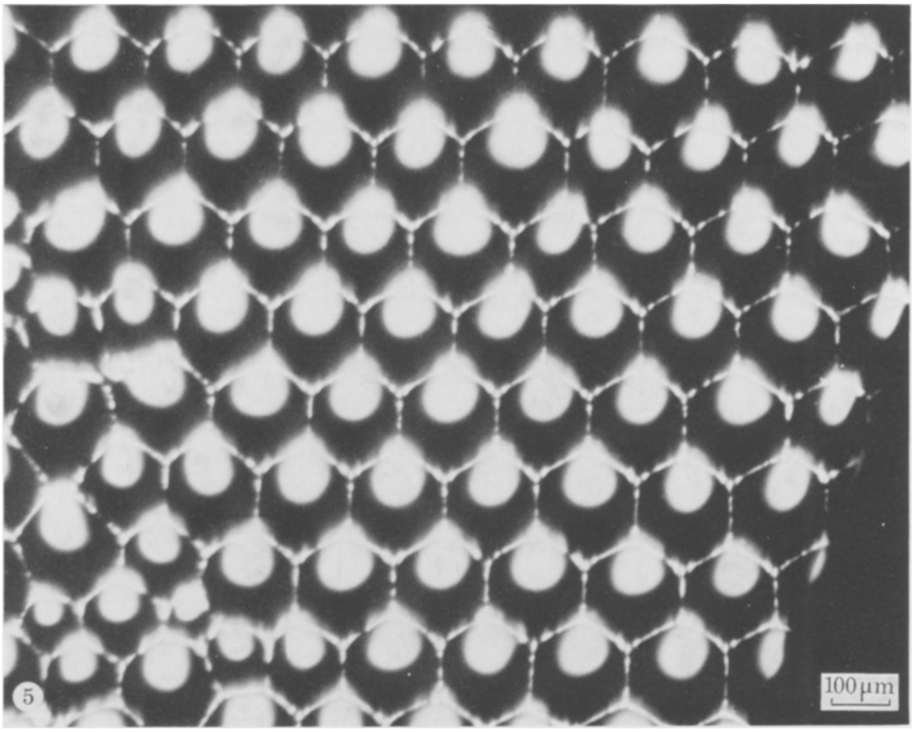

FIGURE 4. Closed circles of type II electroconvection cells, each circle defining a type III cell. At this stage each II cell is discharging continuously. Quaker State motor oil S.A.E. 30, 13 μm thick, 0 °C. 16 μA/cm².

FIGURE 5. Type III electroconvection cells at high population density have regular hexagonal boundaries. Each bright spot in the cell boundary is the remnant of a II cell discharging continuously from the free surface to the underlying conducting support. Mixed aromatic oils, $10^{-3}\,m^2\,s^{-1}$, 13 μm thick, 23 °C, 516 μA/cm². Examples were selected for figures 2 to 5 which best illustrated the stage of development in preference to showing the sequence in a single liquid.

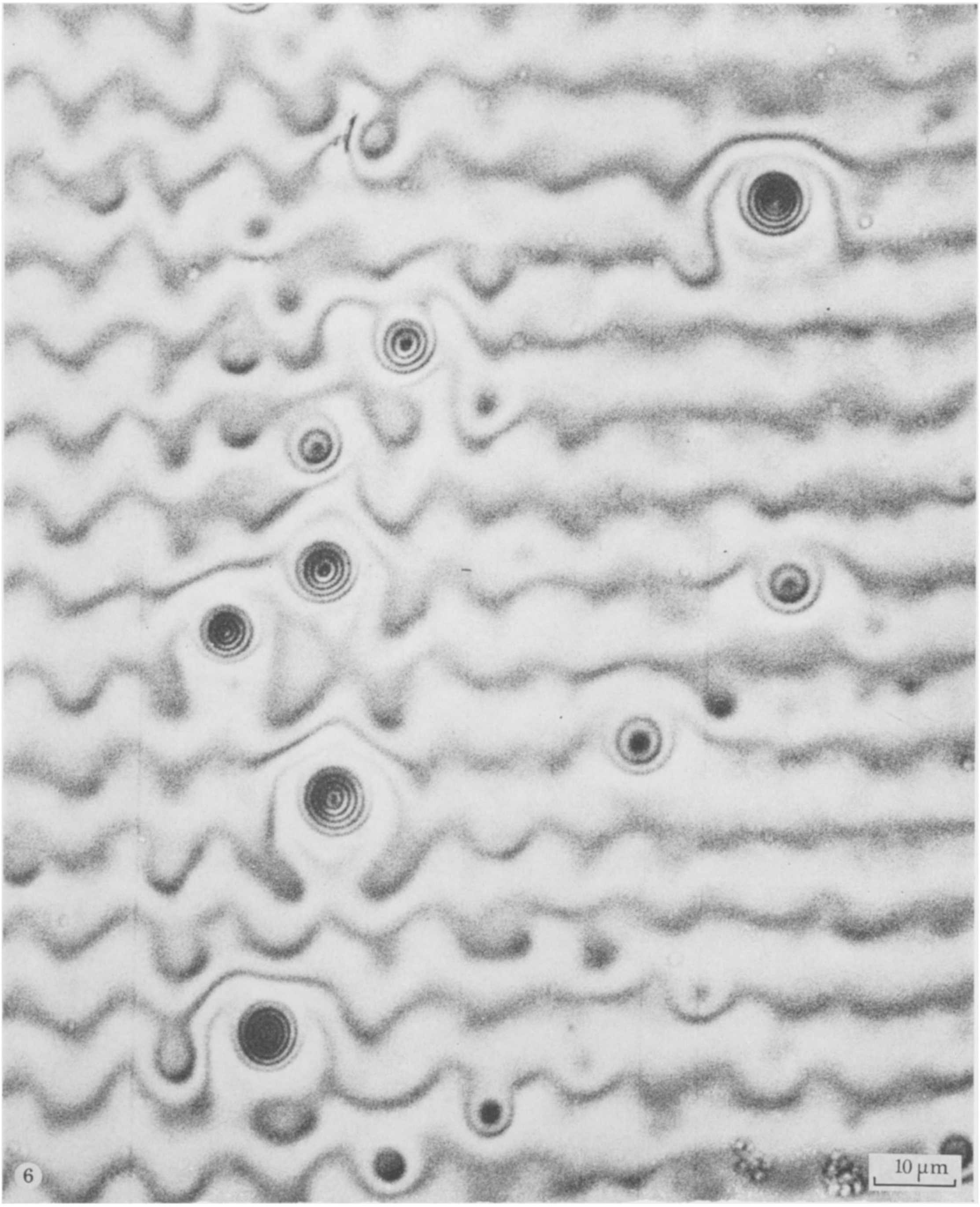

FIGURE 6. Interference micrograph of type II electroconvection cells frozen in solid poly-diphenylsiloxane 7 μm thick. The sample was surface charged by electrons in a thermoplastic recorder (1000 μA/cm² for $\frac{1}{60}$ s), thermally softened for about 2 s to allow deformation, resolidified, aluminized to 60 % transmission, and illuminated with 540 nm light.

It is possible that this phenomenon was actually II cell formation and that the accompanying ring vortex of fluid could not be observed owing to the physical arrangement of the experiment. If this is true then II cell cones should have a limiting vertex angle of 98.6° immediately before discharge.

An attempt was made to measure the vertex angle of II cells in the following way. Figure 6, plate 23, shows a group of II cells formed and captured in solid poly-diphenylsiloxane by the thermoplastic recording technique (Glenn 1959). Here a stream of electrons in a vacuum chamber lands on a solid dielectric thermoplastic surface which is later heated momentarily to a molten state then quickly cooled to freeze in place the deformations caused by the deposited charge. It is electromechanically equivalent to the corona technique and the same electroconvection cells are observable. The presence of raster lines however interferes somewhat with I and III cells because of their size and distributed electrical fields but not with II cells because of their concentrated electrical field and because of the special condition that the thermoplastic thickness (7 μm) is roughly equal to the line spacing (10 μm).

The II cells of figure 6 were coated with aluminium to 60% transmission then photographed under an interference microscope using 540 nm light. Measurements on six well-formed II cells indicate that each is a nearly perfect right circular conical depression in the surface about 3.5 μm deep. The vertex angle averages 109° with maximum variations of $+7°$ and $-3°$ in 36 measurements. This value compares favourably with Taylor's limiting value of 98.6° considering that equilibrium deformation is not likely to be fully attained in a highly viscous system within the 2 s available before resolidification. This is consistent with the measured angle being slightly larger than Taylor's value. The good agreement is taken to be experimental support for the suggested structure of II cells.

The types and behaviour of electroconvection cells reported here differ substantially from those of Avsec & Luntz (1937). The A. & L. 'b' cell appears identical to the III cell. The A. & L. 'a' cell has a conical shape similar to the II cell but polygonal boundaries were not observed here under any condition so it is unlikely that A. & L. observed II cells. A. & L. did not observe II cell discharge nor did they observe I cells due probably to the greater liquid thickness used in their experiments.

Strings of II cells in random wrinkled shapes (figure 3 without the discharges) have appeared in the literature on several occasions without being identified as electroconvection cells. In each case solid polymer layers have been given a surface charge followed by sufficient heating to allow deformation. Gundlach & Claus (1963) called the phenomenon 'frosting'. Thomas (1951) described it as 'heat developed Lichtenburg figures', while Nicoll (1964) used the term 'crazing'.

The term 'convection cell ridge pattern' is suggested for future use as being more accurately descriptive of the phenomenon.

Despite the great difference in scale a similarity is noted between II cell structure and that of an atmospheric tornado.

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