

Computation of current distribution in electrodeposition, a review

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This paper reviews the research literature that has appeared since 1980 on computer calculations of current distribution in the field of electrodeposition. Key contributions and general trends are identified, with particular emphasis given to applications in the electronics industry. The survey reveals how numerical models have provided the technology of electrodeposition with general understanding, predictive power, and the capability of optimizing deposit uniformity. Anticipated developments for the nineties are discussed.

Introduction

The importance of electrodeposition as a fabrication technology in the electronics industry is large and growing. With the current trends toward miniaturization, cost competitiveness, and high-performance packaging, electrodeposition has become the dominant

manufacturing technology in many new applications and remains firmly established in others. Key applications of plating that have achieved major importance in the eighties are tape-automated-bonding (TAB) packages, dual-in-line packages (DIP), thin-film magnetic-recording heads, electromagnetic interference (EMI) shielding of system enclosures, and integrated-circuit packaging by controlled collapse chip connection (C4). Applications of longer standing that remain vital are printed-wiring boards, contacts, connectors, and corrosion protection. A good appreciation of the breadth of electrodeposition technology in use in the electronics industry can be gained from the proceedings of a recent symposium [1], and the importance of plating to electronic packaging is conveyed in recent monographs [2-4].

In nearly every application of electrodeposition, the pursuit of deposit *uniformity*, especially thickness uniformity, has been a technical imperative. However, nowhere has uniformity been as important as in the electronics and computer industries, where the most stringent requirements on electrodeposition have been imposed. The success of electrodeposition in these industries has depended largely upon efforts to achieve uniform films of a metal or alloy in spite of difficulties presented by lithographic patterns, irregular surface topographies, narrow holes, large substrates, and resistive seed layers.

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In general, a uniform electrodeposit is achieved when the current density is evenly distributed over the electrode surface. Since electrodeposition is a Faradaic charge-transfer reaction, the rate of deposition depends upon the current density. In the absence of any interfering reaction, the relationship is Faraday's law:

$$M = \frac{i}{nF} \frac{A}{\rho}, \quad (1)$$

where M is the deposition rate in cm/s, i is the component of current density normal to the electrode surface in A/cm², n is the number of electrons transferred per metal atom discharged, F is Faraday's constant (96 487 C), A is the atomic weight of the metal in grams, and ρ is the density of the metal in g/cm³. In this case, the thickness distribution is proportional to the normal current-density distribution for deposits that are very thin compared to the size of shape features on the substrate. When there is a competing reaction, such as hydrogen gas evolution, the current efficiency of plating (i.e., the fraction of the current density that participates in the electrodeposition reaction) generally varies with current density; despite this complication, thickness uniformity is nearly always best achieved when the current density is uniform. When two or more species are deposited simultaneously from the same solution to form an alloy, the compositional uniformity of the deposit may be at least as important as the uniformity of the thickness, especially in the fabrication of magnetic devices. Again, however, composition generally depends directly on current density, as do other deposit properties such as roughness and intrinsic stress. In most cases, therefore, the challenge of deposit uniformity can be posed as a problem of current-density distribution: How can one distribute the electrolytic current evenly over the surface of the cathode?

The nature of the electrodeposition process is such that there exist strong tendencies for the current distribution to be nonuniform. The best-known effect of this type is due to nonuniformities in the electric potential field that result from the shape and geometric configuration of the electrodes. A nonuniform current distribution can also arise when the following two conditions are met. First, the deposition reaction must be *influenced* by the rate of mass transfer of one or more active species. Second, the rate of mass transfer must be *spatially nonuniform* over the electrode surface. The latter condition can apply when the agitation of the electrolyte is uneven or when electrode shape features of size comparable to the diffusion-layer thickness give rise to nonuniform diffusion. In many cases, the qualitative understanding of such effects has been established, and recent progress has been mainly in the judicious application of scientific

knowledge in engineering analysis of practical systems. This paper focuses on advances in the use of quantitative mathematical models to determine current distribution and, ultimately, to achieve optimum deposit uniformity.

It can be said that the capability of precise prediction of current-density distribution has been one of the foremost achievements in what Carl Wagner has called "the science of electrochemical engineering" [5]. It is one of the advances that has elevated electroplating from an empirical art to a sophisticated technology.

The purpose of this review is to summarize the significant advances in the field made during the eighties. The large body of progress made in the seventies and earlier has been well summarized in a 1982 review paper by Prentice and Tobias [6]. Unlike that review, this work focuses on electrodeposition rather than on the entirety of electrolysis. Work from related fields, such as etching and corrosion protection, is covered only to the extent that its treatment of current-distribution calculations is likely to have an impact on electrodeposition.

One can observe certain general trends in the treatment of current-density calculations in the eighties. Not surprisingly, the emphasis has been increasingly computational and numerical. Analytical treatments are less prominent, and even a number of these rely on numerical evaluation of integral equations. Several methods have been developed for use on personal computers, which continue to grow in power and availability; the pioneering work of Klingert, Lynn, and Tobias [7], performed in 1963 on a state-of-the-art mainframe computer, could be repeated today in less time on a desktop computer.

Unfortunately, the zeal to apply computers to current-distribution problems has produced some "reinvention of the wheel" and failure to build appropriately upon existing foundations. Some analysts have neglected to recognize the parametric dependencies of current distribution pointed out by earlier investigators.

As computers and numerical methods have come of age, attention has shifted somewhat from the computational methods themselves to actual electrochemical problems of interest. Furthermore, there has been a general trend toward specialized application of modeling to specific technical problems.

Classification of current-distribution studies

Possibly the most logical way to categorize studies of current distribution in electrodeposition is in terms of the *scale* of the region of interest. Since it is seldom possible to represent a structure of great complexity, such as a multichip packaging module, in a single geometric model, it is often necessary to decompose the problem into simpler problems, each representing a different length scale. This allows one to determine the essential

characteristics of current distribution on one scale at a time, while disregarding the others. Such an approach implies an eventual reconstruction of the multiscale problem by somehow superposing or merging the single-scale solutions, although this step has thus far escaped attention in the literature. In many cases, the structure of the workpiece quite naturally suggests distinct length scale of interest. For the problem of copper plating of through-holes in multilayer printed-circuit boards, Kessler and Alkire [8] identified three length scales: 1) the *macroscale*, characteristic of the size of the entire board, 2) the *miniscale*, characteristic of the size of individual plated through-holes, and 3) the *microscale*, characteristic of the size of surface roughness. Such a decomposition of the problem is quite appropriate and, indeed, necessary, since it would be virtually impossible to solve for the current distribution over a 50-cm board at the level of detail that treats axial nonuniformity within each of thousands of 500- μm -diameter through-holes. The present work adopts a similar, though enlarged and more general, hierarchy of size scales:

1. *Workpiece scale* characteristic of the entire object undergoing electrodeposition—e.g., wafer, circuit board, or tape.
2. *Pattern scale* characteristic of patterns or regions on the workpiece surface that differ in *active area per unit area* as a result of masking or density of surface features.
3. *Feature scale* characteristic of individual small features such as wiring lines, bonding pads, vias, through-holes, and magnetic recording heads.
4. *Roughness scale* characteristic of microscopic crystallographic roughness, naturally occurring asperities, etc.

While Scales 1, 3, and 4 correspond closely to those outlined by Kessler and Alkire [8] for printed-circuit-board plating, Scale 2, the pattern scale, has received little attention. An example of the usefulness of the pattern scale occurs in the case of a wafer that contains an array of repeating photoresist patterns that correspond to individual chips that will be created when the wafer is diced. As is discussed later, nonuniformities in deposit thickness can occur across each chip pattern as a result of nonuniform density of resist across the pattern.

This paper, which focuses more on the art of fabricating devices than on the phenomenology of electrocrystallization, does not concern itself with the roughness scale. The ample literature on topics of roughness development in electrodeposition (exemplified by two recent contributions that review earlier work [9, 10]) and on phenomena such as leveling [11–13] lies outside the scope of this review. It should be noted,

however, that general methods for calculating current distribution have found use in such studies. It must also be stated that structures fabricated by electrodeposition are steadily shrinking and, in some cases, have already “intruded upon” the roughness scale.

Computational methods for potential-theory problems

Clearly there has been much progress over the last decade in computational methods that are applicable to the class of current-distribution problems described by potential theory. The potential-theory model, a good explanation of which is given by Newman [14], is well established and has been validated by experiment. For the model to be applicable, the electrolyte, bounded by electrodes and insulators, must be of uniform conductivity κ , a condition which is often amply satisfied when the electrolyte is well mixed or when the ions that are participating in the reactions are responsible for only a small fraction of the ionic conductivity of the electrolyte. If this is satisfied, the potential ϕ obeys Laplace's equation within the electrolyte:

$$\nabla^2 \phi = 0. \quad (2)$$

The boundary condition at all insulating boundaries or planes of symmetry is

$$\frac{\partial \phi}{\partial \mathbf{n}} = 0, \quad (3)$$

where $\mathbf{n}$ is a unit vector normal to the surface. The boundary conditions at electrode surfaces contain expressions for electrode kinetics and concentration polarization and take the form

$$\phi = \phi_E - \eta, \quad (4)$$

where ϕ_E is the potential of the electrode itself, and η is the total overpotential. The total overpotential is often decomposed into a kinetic or surface overpotential η_s and a concentration overpotential η_c . The most commonly used expression of electrode kinetics, which relates the surface overpotential to the current density i , is the Butler–Volmer expression,

$$\frac{i}{i_0} = e^{\alpha_a(F\eta_s/RT)} - e^{-\alpha_c(F\eta_s/RT)}, \quad (5)$$

where i_0 is the exchange current density, and α_a and α_c are the kinetic transfer coefficients of the anodic and cathodic components of the electrodeposition reaction. The normal derivative of potential is related to current density by a form of Ohm's law,

$$i = -\kappa \frac{\partial \phi}{\partial \mathbf{n}}. \quad (6)$$

The exchange current density, i_0 , may be taken to depend on the concentration of the electrodeposition ion, c_{M^z} , in the following way:

$$i_0 = i_0^\infty \left(\frac{c_{M^z}}{c_{M^z}^\infty} \right)^\gamma, \quad (7)$$

where γ is a constant, and the superscript ∞ denotes the value at which the concentration is at its bulk value. The concentration overpotential is often represented as

$$\eta_c = \frac{RT}{nF} \ln \frac{c_{M^z}}{c_{M^z}^\infty}, \quad (8)$$

in which n is the number of electrons transferred in the electrodeposition reaction. The potential field, from which the current distribution is determined, must satisfy Equations (2)–(8). When the total overpotential is assumed to be zero, the result is referred to as the *primary distribution* and depends solely on the geometry of the problem. When kinetic limitations are considered [as in Equation (5)] in the absence of concentration variations, the result is called the *secondary distribution*. When concentration variations enter the analysis, the result is known as the *tertiary distribution*, although some have used this term to specify the extreme case in which mass transfer controls the reaction rate to the exclusion of ohmic and kinetic effects. Many analyses of secondary current distribution have treated one of the two limiting cases of the Butler–Volmer rate law, namely the *Tafel* expression, valid at high overpotential,

$$\frac{i}{i_0} = -e^{-\alpha_c(F\eta_c/RT)}, \quad (9)$$

or the *linear* approximation, valid at low overpotential,¹

$$\frac{i}{i_0} = \frac{(\alpha_a + \alpha_c)F}{RT} \eta_s. \quad (10)$$

¹ There has been an unfortunate lack of consensus in the literature on the particular formulation and nomenclature for the dimensionless groups that characterize the boundary-value problem stated by Equations (2)–(6). For either limiting case [Equation (9) or (10)], it has been well established that the current distribution can be characterized by a single dimensionless group, which has been named the Wagner number [15]:

$$Wa = \frac{\kappa}{L} \frac{\partial \eta_s}{\partial i} \Big|_{i=i_{AVE}},$$

where i_{AVE} is an average current density and L is a characteristic length. As Newman [14] has shown, however, three groups are required to specify the solution when neither the Tafel nor the linear limit applies, a fact that has been unrecognized by some. Newman's choice of the trio J , δ , and α_c/α_a [where $J = (\alpha_a + \alpha_c)i_0 FL/RT\kappa$ and $\delta = (\alpha_a + \alpha_c)i_{AVE} FL/RT\kappa$] is not ideal, since this combination fails to collapse to a single group in the Tafel limit and does not honor the naming convention that has become widely recognized in the literature [10, 11, 16–23]. This author prefers an alternative combination of groups adopted by Matloz et al. [19], Wa_L , Wa_T , and α_c/α_a , where Wa_L and Wa_T correspond to the Wagner number evaluated at the linear and Tafel limits:

$$Wa_L = \frac{RT\kappa}{(\alpha_a + \alpha_c)FLi_0} \quad \text{and} \quad Wa_T = \frac{RT\kappa}{\alpha_c FLi_{AVE}}.$$

For want of such a standard nomenclature, groups identical to the Wagner number or its reciprocal have been variously referred to in the literature as P [8], ξ [24], j^* [25], k_c [26], β [27], Λ [28], and k_c/A [29]. In this review, the Wa_T – Wa_L nomenclature is used when applicable.

As many authors have pointed out, the key features that render many current-distribution problems intractable by analytical methods are the nonlinearity of the η - i dependence [Equation (5)] and the irregular shapes often assumed by the electrodes and insulating boundaries. The earliest known numerical computation of electrochemical current distribution [7] employed a finite difference model (FDM). The seventies saw widespread use of FDM and the introduction of finite element methods (FEM) to current-distribution problems. FDM and FEM are sometimes called “domain methods,” since the boundary-value problem is posed, discretized, and solved on the entire domain. In the eighties, the domain methods continued to see progress, while a class of “boundary methods” began to find use. The latter comprise boundary integral equation methods (BIEM) and boundary element methods (BEM); both are characterized by a recasting of the boundary-value problem entirely on the boundaries of the domain by manipulations such as Green's identities. With the problem posed entirely on the boundaries, the effort of defining the discretized geometry is greatly reduced, and related advantages, such as ease of handling moving-boundary problems, result [18, 26, 30]. It should be pointed out that the FDM, FEM, and BEM methods were developed in the fields of civil and mechanical engineering and were later adopted for calculation of electrochemical current distribution. More information on these numerical methods can be found in [31–35]. Many investigators have emphasized the advantages of one particular numerical method over another; however, few teams have demonstrated experience with more than one method, and very few direct comparisons between methods have been made on a rigorous basis. Hume, Deen, and Brown [30] have offered a valuable comparison of FEM and BEM for current-distribution problems.

A noteworthy application of the BIEM approach to current distribution in electrodeposition was a 1980 paper by Blue [26]. This paper was remarkably thorough and resourceful, both in its description of the plating of printed-circuit boards and in its approach to the numerical solution of the problem. Based firmly on an earlier, more general communication by the same author on boundary-integral solutions to Laplace's equation [36], the method applies Green's third boundary identity to the derivation of an integral equation. A solution is sought in the form of a piecewise superposition of B-spline basis functions, with special functions employed at singularity points. The electrochemical description includes concentration-dependent Butler–Volmer kinetics and concentration overpotential at both anode and cathode, with vertical variations in the mass-transfer coefficient characteristic of agitation by gas sparging.

Ironically, the plating example chosen exhibited behavior very close to that of the primary current distribution. The efficiency and simplicity of the BIEM approach were clearly demonstrated.

Two investigations using BEM appeared one month apart in 1984, both drawing from BEM methodology published by Brebbia and Walker [35]. Bialecki, Nahlik, and Lapkowski [37] used BEM to calculate the primary current distribution for a configuration of parallel-plate electrodes inside an insulating cylinder. Notably, they avoided placing nodes along two symmetry planes by selecting Green's functions that satisfied these boundary conditions as well as Laplace's equation. The other team, Hume, Deen, and Brown [30], conducted an excellent investigation of patterned electrodeposition, the results of which are described subsequently under the heading *Current distribution on the feature scale*. Thorough attention was given to the issues of solution error and mesh convergence; with quadratic boundary elements, the order of convergence was shown to be roughly quadratic for the root-mean-square solution and cubic for the overall flux balance. The work involved a moving-boundary simulation of electrode shape change. The authors followed this investigation with a study of moving-boundary potential problems, not limited to electrolysis, in which BEM and FEM were systematically compared. For many problems, FEM was found to be competitive with BEM in accuracy and efficiency, and emphasis was placed on the extendability of FEM codes to partial differential equations that cannot be treated by BEM because no Green's function can be found. However, BEM was judged to be superior for problems in which boundary motion is proportional to the normal derivative of the potential. Electrodeposition problems fall into this category.

Another group that made early use of BEM to simulate electrodeposition was Deconinck, Magetto, and Vereecken [18]. Their numerical approach was also based on that of Brebbia and Walker [35] and used a Newton-Raphson iteration in conjunction with a direct matrix solver. The advantages of BEM for problems of current distribution and electrode shape change were illustrated with several relevant examples.

Cahan, Scherson, and Reid [38] developed an implementation of the BIEM approach based on an iterative solution of the integral equations rather than on a matrix formulation. Nonlinearities stemming from the overpotential boundary conditions were handled in the same iterative loop. The memory requirement of this method is small, permitting implementation on personal computers. The method was claimed to have high stability, accuracy, convergence rate, and computational speed, but direct comparisons to other methods were not reported. A moving-boundary implementation of the

method was used to simulate shape change on an electrode with a square corner.

Current distribution at the workpiece scale

Parn et al. [39] applied numerical models to aid in "jigging," which is the advantageous spatial arrangement of the workpiece, counter electrodes, insulating walls, and auxiliary electrodes. Both FEM and BEM were used to predict current distribution in an application of hexavalent chromium plating of an irregularly shaped print roll. Simple expressions approximating measured polarization data and current-density-dependent current efficiency were used. In a second example, involving plating of an anvil, the improved uniformity afforded by auxiliary electrodes, current shields, and a specially shaped anode was evaluated. Some experimental validation was reported. Unfortunately, no numerical details were provided, and few conclusions of general consequence were given.

Matlosz et al. [19] conducted a thorough study of secondary current distribution in the Hull cell, a popular diagnostic tool in the plating industry in which the current density is deliberately nonuniform, so that plating performance can be evaluated over a range of current densities. Butler-Volmer kinetics were assumed in the model, and the general dependence of the current distribution on the parameters Wa_T , Wa_L , and α_a/α_c was reported (see Footnote 1 for definitions). The collapse of the three-parameter correlation into a single-parameter dependency in either the Tafel or the linear regime of kinetics was vividly shown. Interestingly, the most uniform current distribution for a given system sometimes occurs at a current density *between* the two limiting cases (Tafel and linear). Results from BEM and FEM agreed well with each other and with thickness data from acid-copper plating experiments.

Shih and Pickering [29] conducted the first direct investigation of three-dimensional (3D) effects in electrochemical current distribution. (3D models for cathodic protection in the field of corrosion were developed earlier [40, 41], but these studies do not seem to have focused on corner and edge effects in a general way.) The analysis was restricted to the case of linear kinetics at a square electrode in an insulating plane, with a counter electrode infinitely far away. Unfortunately, the seminumerical solution method, which involves using Green's theorem to generate Fredholm integrals of the second kind, is not readily extendable to more complicated systems. However, the essential behavior of square electrodes was characterized and clearly presented: Current density is highest at the four corners and lowest at the center; center-to-corner, mid-edge-to-corner, and center-to-mid-edge current-density profiles were shown for various values of the Wagner number.

It has been known for a long time that in the limiting case of the primary current distribution (i.e., $Wa_T, Wa_L \rightarrow 0$), geometric effects may cause the current density at an electrode edge to be infinite or zero [14]. Two recent theoretical studies [42, 43] have focused on the asymptotic approach to such singular behavior. Smyrl and Newman [42] determined the current distribution at a disk electrode in the Tafel kinetic regime ($Wa_T \ll Wa_L$) for high current densities ($Wa_T \ll 1$). Under such conditions, the primary distribution holds over the electrode, except for a small edge region where kinetic effects are felt. The problem was solved by singular perturbation analysis and finite-difference calculation. The edge current density was found to vary inversely with the square of Wa_T . The result is extendable to geometries other than the disk electrode. West and Newman [43] contributed a similar but more general analysis for the edge region of any flat electrode that meets an insulating plane at an arbitrary angle β . Singular perturbation analyses were performed for both Tafel and linear kinetics to yield current distributions in the edge region for various values of β .

Despite the increased prominence of computational techniques in the eighties, studies based on analytic methods have continued to appear in the literature. While these works have generally employed gross approximations and have disregarded the studies based on numerical methods, the results are of compact form and may be of value to practitioners. Popov et al. [44] published a simple approximation of current distribution for plane-parallel electrodes to explain dendrite growth at the edges of the cathode. Klenov [45] presented an analytic approximation for current distribution based on a piecewise-linear representation of nonlinear overpotential expressions. Perakh [46] presented an analytic solution for primary current distribution in a special slotted cell for electrodeposition onto resistive substrates. Pomogaev, Kruglikov, and Nachinov [47] contributed an approximate but compact description of current distribution in terms of an expression for "throwing power with respect to metal," which is related to "throwing power with respect to current" by an analytical treatment involving current efficiency. Nachinov, Vinogradov, and Kruglikov [48] constructed a nomogram for use in predicting the current distribution in a cell of complicated geometry containing one bath, from the measured distribution for another bath and the measured throwing-power indices of both baths.

• *Studies of specialized applications*

Mehdizadeh et al. [49] published a numerical study of the use of coplanar auxiliary electrodes or "thieves" to

achieve uniformity of thickness on planar cathodes, such as resist-patterned wafers. A BEM model of the secondary current distribution with Tafel kinetics was used to solve the multi-electrode problem and to evaluate the thief current at which the thickness uniformity on the workpiece is optimized. The coplanar thief was shown to exert a powerful influence on workpiece uniformity. The sensitivity of the system to Wa_T , thief size, workpiece-to-thief separation, and thief-to-cell-wall separation was evaluated and summarized.

A treatment of the specific problem of copper deposition onto moving sheets of flexible material with substantial seed-layer resistance was published by D'Amico, DeAngelo, and McLarnon [50]. A one-dimensional, steady-state model of the secondary current distribution with linearized kinetics, including ohmic potential drop within the substrate film, was found to match the behavior of the industrial process quite well.

Dimpault-Darcy and White [27] demonstrated calculation of secondary current distribution in cells, such as bipolar electrolyzers, in which the potential of one or more electrodes may not be known *a priori*. Two-dimensional problems with linear kinetics were solved using TOPAZ2D [51], an FEM program originally written for heat-transfer problems, which allows the specification of overpotential boundary conditions at electrically "floating" electrodes.

• *Periodic electrodeposition*

A major contributing factor to the versatility of electrolytic metal and alloy deposition is the capability of modifying the deposit properties by the use of periodic electrodeposition or "pulse plating." Modulation of current or potential has been used extensively in electrodeposition to influence deposit morphology by altering the conditions of mass-transport and nucleation. Pesco and Cheh [52] recently provided a detailed review of the many applications and theoretical treatments of periodic electrolysis. The impact of periodic electrolysis on the time-average current distribution has been studied by several researchers. Early treatments correctly predicted that since peak current densities in pulse plating were higher than the time-average-equivalent dc current densities, the nonuniformity would be larger, according to a Wagner-number argument.

Chin [17] addressed the subject of current distribution in periodic electrodeposition in the following way. A one-dimensional transport/kinetic model was used to predict the relationship between time-average current density and time-average overpotential for rectangular-current-pulse, pulse-reverse, and double-rectangular-pulse electrolysis. Chin suggested that the Wagner number evaluated from the slope of the resulting time-average polarization curve should characterize the current

distribution in periodic electrolysis in the same manner that the Wagner number characterizes the current distribution in dc electrolysis. On the basis of the above argument and the time-average polarization behavior calculated under different conditions, Chin claimed that pulse-electrolysis systems may exhibit either more or less uniformity than their dc counterparts. The case of improved uniformity, however, was reported to apply mainly to anodic electrolysis rather than to electrodeposition.

Popov, Totovski, and Maksimovic [53] presented a rough analytical estimate to interpret experiments in which periodically reversed current control provided higher uniformity than did pulse plating.

An excellent study was carried out by Fedkiw and Brouns [54], who examined current distribution in periodic-voltage electrodeposition on a flat electrode embedded in an insulating plane. The two-dimensional model included laminar-flow convective diffusion, capacitive charging, and simultaneous hydrogen evolution. Mass-transport effects entered through the concentration-dependent Butler-Volmer equation and the concentration overpotential for the metal-deposition reaction. The stationary-state solution was found by iteratively coupling Laplace's equation for potential to the boundary-layer form of the mass-conservation equation. The system was characterized by ten dimensionless groups. Predicted nonuniformities were expressed as deviations from the equivalent dc distribution, with care taken to select a fair basis of comparison between dc and periodic electrolysis: The charge passed by the metal-deposition reaction was equivalent in the two cases. It was shown that sinusoidal voltage control can result in either an increase or a decrease in uniformity, depending on the transport and kinetic parameters and on the applied voltage. It was also shown that, with all other variables held constant, uniformity improves with increasing pulse frequency.

Wan and Cheh published treatments of the current distribution on a rotating-disk electrode (RDE) under galvanostatic [55] and potentiostatic [56] pulsed electrolysis. The axisymmetric model used in both works included concentration-dependent Butler-Volmer kinetics, concentration overpotential, and a rigorous treatment of convective diffusion. An orthogonal collocation method was employed in an iterative scheme to obtain the full transient solution. Predicted current-density profiles matched acid-copper deposit-thickness profiles measured by cross-sectioning. It was reported that nonuniformity increases with current density and that the surface concentration of the reacting ion is higher at shorter pulse times and duty cycles.

A treatment of current distribution in periodic electrodeposition was applied to the specific problem of

through-hole plating of printed-circuit boards by Pesco and Cheh [57]. This work is described subsequently under the heading *Through-holes in printed-circuit boards*.

The review paper by Pesco and Cheh [52] considered current-distribution effects in periodic electrodeposition and drew heavily from earlier work on tubular electrodes [57] and on the rotating-disk electrode [55].

Finally, Chin, Vilambi, and Sunkara [16] reported on a current-distribution study pertaining to a proposed technology for "selective" or spatially localized electrodeposition based on pulse plating. The method relies on periodic electrolysis to deposit metal at very high current densities, so that a highly nonuniform distribution of metal can be achieved deliberately. The authors invoked the assumption that the time-average current distributions for periodic and dc systems are equivalent provided that the time-average polarization curves are identical. Measured time-average current-voltage behaviors for gold and copper plating baths were entered as boundary conditions in a dc secondary-current-distribution model (based on FEM and orthogonal collocation). Some discrepancy between experimental thickness profiles and the predicted behavior was reported. The experiments did show a slight increase in nonuniformity as a result of pulse plating.

• *Measurement errors associated with current distribution*

Spatial models of current and potential distribution are valuable not only for their ability to describe deposit uniformity but also for the insight they can provide into the interrelation of the potential field and various measurement techniques. Two recent works have made use of numerical potential-field models to investigate errors in measurements of electrode overpotential by the use of Luggin capillaries. Tokuda et al. [23] conducted a remarkably thorough investigation of Luggin probe measurements in which an insulating annular capillary tube is either held with its tip near a polarized planar electrode on the solution side (as in conventional practice) or passed through the electrode from the back side, so that its tip is flush with the active electrode surface. The axisymmetric secondary-current-distribution problem was solved by Galerkin FEM with quadratic quadrilateral elements. A complete analysis of the relation between true and apparent overpotentials was given in terms of Wagner number, probe-to-electrode spacing, and capillary-wall thickness. For convenience of application, the results were expressed not only as potential-field contour plots and current-distribution curves, but also in the form of algebraic approximations. The study confirmed that the potential measured by a conventional Luggin probe can be regarded as the sum of

the true overpotential and the ideal potential drop as long as the probe is at least one probe diameter away from the electrode surface. It was also shown that back-side capillary measurements consistently exaggerate the true overpotential.

Landau, Weinberg, and Gileadi [58] conducted a similar investigation, which compared various placements of the Luggin probe in parallel-plate cells. (Unfortunately, this team disregarded the work of Tokuda et al. [23].) The axisymmetric potential problem with Tafel kinetics was solved by CELL-DESIGN² [59], a commercially available, interactive software package, based on FDM, for computer-aided design of electrochemical cells. The theoretical study concluded that the smallest measurement error is introduced when the capillary consists of an uninsulated hole through the electrode accessed from the back side. The authors also showed, as had Tokuda et al. [23], that the other method of back-side access, in which the wall of the hole is an insulator, is particularly disadvantageous. Other recommendations of the investigation are to avoid placing the probe behind one electrode outside the interelectrode gap and to subtract the ohmic drop by calculation when possible, rather than reducing the ohmic drop by placing the probe very near the electrode surface.

West and Newman [60] carried out BEM calculations of potential distribution to determine the extent to which nonuniform current distribution causes error in the measurement of kinetic data on a rotating-disk electrode. The treatment, valid for Tafel kinetics in the absence of concentration variations, followed an earlier analysis for linear kinetics [61]. The results were interpreted in the practical form of equations that yield corrected values of the exchange current density and the transfer coefficient from their apparent values and in terms of recommended conditions for kinetic measurements.

Current distribution at the pattern scale

Current-distribution effects at the pattern scale are relevant chiefly to applications of electrodeposition in the electronics industry in which a lithographic pattern of small features is present on the workpiece surface. Nonuniformity effects arise when different regions of the surface possess different *densities* of patterning. For example, neighboring zones of high and low photoresist density in a wiring pattern can give rise to uneven deposit thickness from one zone to another, even if an otherwise uniform current distribution has been achieved on the workpiece scale. This effect has been noted in the literature by Romankiw et al. [62], by Horkans and Romankiw [63], and most recently by Yung and

Romankiw [64]. The phenomenon, which can be called the *active-area-density effect*, is unique to electrodeposition as compared to other metallization processes used in the electronics industry, such as evaporation, sputter deposition, chemical vapor deposition, and electroless deposition. Mehdizadeh et al.³ [65] developed a theoretical description of patterned electrodeposition that enables one to solve problems that involve the active-area-density effect by the use of numerical algorithms developed for ordinary problems of secondary current distribution. Two neighboring zones in the pattern are approximated as continua, each with a different *active-area density*, i.e., active area per unit superficial area. The overpotential expression applicable to each zone is modified to reflect the difference between the true or *surface* current density and the overall or *superficial* current density. The resulting secondary-current-distribution problem is solved numerically. The results show that in the Tafel regime, the pattern-scale nonuniformity depends on two parameters: a Wagner number based on the average superficial current density, and the ratio of active area densities between the two zones. The Wagner number depends in the usual way on the characteristic length, which in this case reflects the sizes of the zones, and the model predicts that uniformity is greatest when the zones are small. The theory applies not only when the electrode's active-area density is decreased by resist coverage but also when the active-area density is increased by topographic features such as through-holes.

Current distribution at the feature scale

Nonuniformity effects that occur at the scale of small features on the surface of a patterned substrate may be qualitatively different from those across larger regions. In the present categorization, a "feature" can range in size from a 500- μm through-hole [66] to a 0.2- μm -wide element in the gold absorber pattern of an X-ray lithography mask [67]. Features at this scale can be comparable in size to the thickness of the concentration boundary layer; accordingly, treatments of mass transfer that assume a mass-transfer boundary layer that conforms to the electrode profile may be invalid. As dimensions shrink, the importance of ohmic potential variations diminishes. However, nonuniformities in mass transfer, due to geometric effects such as spherical diffusion, can give rise to strong deposit nonuniformities by causing local variations in concentration overpotential and concentration-dependent kinetics. Furthermore, at this small scale the shape of the electrode can be changed significantly by the deposition of films that could be

² CELL-DESIGN is a registered trademark of L-Chem, Inc., 13909 Larchmere Blvd., Shaker Heights, OH 44120.

³ S. Mehdizadeh, J. Dukovic, P. C. Andricacos, L. T. Romankiw, and H. Y. Cheh, "A Model for the Influence of Substrate Patterning on Electrodeposit Uniformity," submitted to *J. Electrochem. Soc.* (1990).

considered negligibly thin at larger scales. Accordingly, in the fabrication of microelectronic structures, the capability of simulating the shape evolution with a moving-boundary model is highly advantageous.

An important early treatment of electrodeposition at the feature scale was conducted by Alkire, Bergh, and Sani [68]. This work considered secondary current distribution and electrode shape change in electrodeposition onto an alternating pattern of coplanar conducting and insulating stripes. In this first application of FEM to moving-boundary problems of electrochemical potential, profile-growth simulations exhibiting lateral overgrowth of the deposit over the insulating stripes were conducted for different values of applied potential and a linear polarization parameter. The authors acknowledged that the moving-boundary scheme and treatment of singularities were simple, but also made thoughtful remarks on possible refinements and future models of this type.

Alkire and Reiser [24] built upon the above treatment to compare predicted and experimental results for acid-copper deposition onto an array of parallel stripes. A different polarization parameter, based on Tafel kinetics, was shown to characterize the shape-change behavior. On the scale of the experiment (a 0.79-mm unit of symmetry), lateral overgrowth approximately matched vertical growth at the electrode edge, and the measured vertical nonuniformity slightly exceeded the predicted value.

An analysis by Hume, Deen, and Brown [30] focused on electrodeposition of features "small relative to the concentration boundary-layer thickness" that are defined by polymeric masks that are not negligibly thin. Concentration effects were treated by solving the diffusion equation within an assumed stagnant layer that is thicker than the mask material and the growing deposit. Potential variations due to ohmic effects were ignored, and the potential field was not solved. The degree of nonuniformity of the deposit was found to depend mainly on a parameter ξ that is proportional to the ratio of the current density to the limiting current density. At low ξ the growth is even, whereas for high ξ the growth is faster near the resist wall, because of metal ions supplied from above the resist structure. When the diffusion boundary layer is made thinner, this extra supply and the resultant nonuniformity diminish. In general, deposition at rates below 40 percent of the limiting current is not significantly nonuniform. When nonuniformity does occur, its magnitude accelerates as the deposit grows. The BEM with isoparametric, quadratic elements was shown to be well suited to shape-change problems of this type.

Menon and Landau [69] presented a specialized model of current distribution that is applicable to stagnant

electrolytes or electrolyte layers. The treatment was developed for binary electrolytes and accounts for transient diffusion and migration. The solution was obtained on a personal computer by a modified alternating-direction implicit (ADI) implementation of FDM. In this system, the fraction of the current density due to migration varies with time and position in the cell. It was found that the diffusion resistance typically causes nonuniformity to increase with time.

- *Through-holes in printed-circuit boards*

Perhaps the most familiar problem of electrodeposition uniformity of small features is the plating of through-holes on printed-circuit boards. In this technology, one seeks to metallize as uniformly as possible the interior walls of holes that are approximately 0.5 mm in diameter and 6 mm in length [66]; the latter dimension corresponds to the thickness of the board. The issue of nonuniformity has become more challenging with time, as increasingly long and narrow holes have been required. A good review of the technology and of the various published studies of uniformity has been provided by Yung, Romankiw, and Alkire [70].

The benchmark treatment of the through-hole problem appeared in a pair of papers from 1976 by Kessler and Alkire [8, 71], based in part on a model of tubular electrodes by Alkire and Mirarefi [72]. Fully developed laminar flow within the hole is assumed, and the convective diffusion equation for the cupric ion is coupled to a one-dimensional charge-balance equation for potential. Two dimensionless groups that characterize the problem are identified: $\xi_{T\kappa}$, a Tafel polarization parameter that reflects the relative importance of ohmic and kinetic resistance, and N , a dimensionless, average limiting current density, which reflects the relative importance of ohmic and mass-transfer limitations.

Ben-Porat, Yahalom, and Rubin [73] developed a similar model of somewhat greater complexity by including the transient effect of finite ohmic resistance in the substrate layer. Periodically fluctuating fluid flow was also included in the model.

Sullivan and Middleman [74] described the through-hole system with a model that neglects ohmic potential drop and assumes that the plating rate is entirely controlled by mass transfer. Since many would consider both assumptions inappropriate to the problem of through-hole plating, the results of the study are questionable, except as a confirmation that plating rates would be severely limited in the absence of convection. A follow-up paper by Middleman [75], based on the same assumptions, compares unidirectional and periodically reversed flows. The latter are found to be more effective, but the fact that only mild convection is required to remove any influence of transport resistance

(as emphasized by Lanzi and Landau [76]) is not recognized.

Alkire and Ju [77] treated the somewhat specialized problem of a through-hole agitated by a coaxial impinging jet. Mass-transfer behavior under both submerged and unsubmerged jets was characterized theoretically and confirmed experimentally by the use of sectioned electrodes and the reduction of ferricyanide to ferrocyanide as a mass-transfer-indicating reaction. It was found that for current below one fifth of the limiting current, no mass-transfer influence is felt.

Lanzi and Landau [76] performed individual calculations of the mass-transfer-limited current distribution for oscillating fluid flow inside a through-hole and of the secondary current distribution under Tafel kinetics. They concluded that, under typical industrial conditions, it is the ohmic resistance rather than the mass-transfer resistance that limits uniformity. Measures that can be taken to improve uniformity are cited, such as raising bath conductivity or polarization resistance, or changing the system geometry to reduce the ratio L^2/R , where L and R are the length and radius of the through-hole.

Lanzi et al. [78] conducted a theoretical investigation of the possibility of improving through-hole plating uniformity by modifying the electrode-kinetic behavior with additives. Electrode-kinetic curves from three different bath formulations were entered as boundary conditions in a one-dimensional model of secondary current distribution, based on several approximations. It was concluded that minor changes in kinetic behavior may profoundly affect through-hole current distribution.

Pesco and Cheh [57, 79] recently contributed two theoretical studies on the subject of through-hole plating. The first [79] considered the influence of fluid flow within the hole. They rigorously solved the coupled mass-transfer and ohmic-potential problems, making use of separation of variables, combination of variables, Duhamel's theorem, the method of Acrivos and Chambre, and a modified orthogonal-collocation method. The result for a stagnant electrolyte reveals that current densities higher than 1 mA/cm^2 cannot be achieved without fluid flow. The solutions for a unidirectional flow of 25 cm/s show that for length-to-diameter ratios $5:1$ and $10:1$, the chief limitation to current density inside the hole is ohmic (as shown by Lanzi and Landau [76]), although the current density may approach its mass-transfer limit at either end of the hole.

The second study by Pesco and Cheh [57] considered the use of periodic potential control in through-hole plating. The problem of transient mass transfer under developed laminar flow coupled to the potential problem via the concentration overpotential and concentration-

dependent kinetic overpotential was solved by an elaborate method involving orthogonal collocation and implicit integration over each time step. Periodic electrolysis was shown to increase the nonuniformity over the dc case. Calculations that disregarded concentration effects showed that deposition with periodically reversed current can improve uniformity with respect to the dc case under some conditions.

Finally, as part of a mainly experimental investigation, Yung and Romankiw [64] developed a model that encompasses both the through-hole and the surface of the board. It was generally found that the deposit thickness on the board decreases near the mouth of a through-hole and further decreases within the hole. Conclusions regarding the factors that limit uniformity and suggested measures for overcoming these were given.

• *Special applications*

Current distribution plays a pivotal role in the special technique of "selective" plating, in which a deliberately high nonuniformity is generally sought. Two investigations on the use of jets of electrolyte to achieve selective deposition were conducted by Alkire and Chen [80] and by Alkire and Ju [25]. Both investigations were accompanied by theoretical treatments of the current distribution. The first paper [80] dealt with unsubmerged, circular jets, as from a round nozzle. The model included ohmic conduction within the electrolyte stream of the impinging jet, mass transfer, and charge-transfer overpotential for multiple electrode reactions. Factors influencing the spatial selectivity of the deposition rate were identified. The interdependence of model and experiment and the value of mathematical models in the design process were emphasized. The second paper [25] treated two-dimensional jet flow, as from a slot, which was modeled in a similar way. It was found that flow conditions exert an especially strong effect at higher current densities, at which mass-transfer effects are important. Dimensionless groups useful for scaling up the apparatus were identified, and the potential engineering value of the model was discussed.

Activity outside electrodeposition

Methods and approaches for calculating current distribution in electrodeposition systems have often come from other areas. Several recent noteworthy advances are pointed out in this section. Sautebin and Landolt [11] conducted a moving-boundary FEM simulation of anodic leveling of a triangular surface profile, with concentration effects handled by a stagnant-diffusion-layer approximation. A succeeding study by Clerc and Landolt [12] examined the dependence of FEM accuracy on the size of elements and time steps. Alkire, Reiser, and Sani [81] rigorously treated convective diffusion in a

dissolution problem involving recirculating flow inside cavities; FIDAP,⁴ a commercial FEM program, was used, and its extendability to more complicated systems was emphasized. The same approach to modeling fluid flow and convective transport was employed by Alkire and Deligianni [82] in a study of anisotropic etching. Finally, two papers on cathodic protection technology in the field of corrosion illustrated the use of three-dimensional potential-theory models with nonlinear, time-dependent kinetic expressions that reflect surface fouling of steel structures in contact with sea water. Gartland and Johnsen [41] used a commercial BEM program called COMCAPS⁵ with rectangular elements; the potential was assumed constant over each element. Strommen et al. [40] reviewed advances in models for cathodic protection of offshore structures. The use of tubular boundary elements to describe three-dimensional structures and the use of digitized empirical polarization data were illustrated. The advantages of BEM for such applications were enumerated.

Anticipated developments in the 1990s

The nineties will certainly see continued progress in electrodeposition-distribution modeling, with important changes driven largely by the electronics industry. Electrodeposition is likely to remain a vital fabrication process for reasons of technical viability and cost, especially for packaging, where technical and cost-related demands have already begun to surpass those of integrated-circuit fabrication. The demands for precision and uniformity will certainly intensify as features and tolerances continue to shrink. In general, it is probable that the better-established models will find widespread use and that research will extend the scope of models to encompass more subtle and realistic effects.

The implementation of current-distribution models as computer-aided engineering software is certain to become widespread, as powerful workstations continue to proliferate. The numerical details of such commercial interactive programs will be invisible to the user occupied with the need to optimize a manufacturing process. The sophistication of *electrodeposition process models* will approach that of *semiconductor process models*, such as SUPREM III⁶ [83] and BICEPS⁷ [84]. The latter category, in turn, is constantly being improved, with the

aim of achieving the geometric realism and predictive capability of *device-performance models*, such as FIELDAY⁸ [85] and CADDETH⁹ [86]. Precise simulation of the shape evolution of electrodeposited features will be required in the context of 3D solid models for multistep component fabrication. Such multistep process models, in conjunction with device-performance simulation, will enable optimized design for manufacturability as well as the exploration of new designs and processes.

Research areas in electrodeposition-distribution modeling will be linked to advances in the science of electrodeposition itself and will include the effects of chemical-additive systems on feature-scale current distribution, alloy deposition with anomalous kinetics, and the evolution of microcrystalline morphology. All enduring progress in these areas will be predicated on conclusive experiments, as the predictive power of numerical models can surpass neither the correctness of the physical description nor the precision with which the process parameters are known.

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⁴ FIDAP is a registered trademark of Fluid Dynamics International, Inc., 1600 Orrington Ave., Suite 400, Evanston, IL 60201.

⁵ COMCAPS is a computer program developed by SINTEF, Division of materials and processes, 7034 Trondheim-NTH, Norway.

⁶ SUPREM III is a computer program developed by the Integrated Circuits Laboratory, Stanford University, Stanford, CA 94305.

⁷ BICEPS is a computer program developed at AT&T Bell Laboratories, 600 Mountain Ave., Murray Hill, NJ 07974.

⁸ FIELDAY is a computer program developed by IBM, General Technology Division, Essex Junction, VT 05452.

⁹ CADDETH is a computer program developed by Hitachi Co., Ltd., Tokyo, Japan.

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