Simulation of Electrocodeposition

Jaesung Lee and Jan B. Talbot
Chemical Engineering Program
University of California, San Diego
9500 Gilman Dr., La Jolla, CA 92093-0411

Metallic matrix composite films are produced by occlusion of particles during electroplating. This process has many advantages in industrial applications due to the uniformity of deposition, even for complex shapes, and the avoidance of high temperature and high pressure processing. Our experimental research to date has been primarily focused on electrocodeposition under controlled hydrodynamics using a rotating cylinder electrode or impinging jet as a function of process variables, such as current density and bath composition, for copper-alumina composites [1]. However, the physical phenomenon of electrocodeposition is not well understood [2]. Even with a primary current distribution, there has not been a study to show the entire entrapment process. The objective of this study is to first simulate the incorporation of a particle by the growing metal film for primary, secondary, and tertiary current distributions in a stationary fluid and then with the influence of convection.

As a first step, the particle incorporation process in a stationary fluid was numerically studied assuming that the effect of lateral migration of the fluid between a particle and the electrode is negligible. The growth of an electrodeposited film along the electrode is governed by the local current density that is affected by the presence of a non-conducting particle. Since the potential gradient is negligible in a supporting electrolyte, the current density depends on the concentration gradient of metallic ions. One of the goals is to investigate the resistance variation in the presence of an inert particle with primary, secondary, and tertiary current distributions.

In order to simulate the particle incorporation process with a tertiary current distribution, a uniform average current density is initially assumed, and the gradient of concentration is computed. The surface concentration is obtained by solution of the Laplace equation for concentration, and the surface overpotential is given by the Bulter-Volmer equation. The total overpotential is used as a boundary condition on the metallic surface to obtain the potential field. Then, the current density can be computed from the potential gradient. The procedure is iterated until the difference between guessed and computed current density converged to a given tolerance [3,4]. When the current density is computed, the moving metallic interface is tracked implicitly by the level set approach that enables the simulation of the entrapment process around the non-conducting particle [5]. The PLTMG (Piecewise Linear Triangle Multi Grid Package) developed by Bank [6] was used to solve the Laplace equation for concentration and potential fields.

The evolution of the metallic surface is affected by the presence of a non-conducting particle. Figure 1 shows the variation of current density with the particle's position, where H (=y/a) is the normalized distance from the electrode and a is the particle radius. The presence of an inert particle significantly affects the current density along the flat electrode as a particle moves in the vicinity of the electrode, which in turn affects the evolution of the metallic interface. Figure 2 represents the variation of resistance δR induced by an inert particle for a primary current distribution, where the resistance variation is defined as the ratio of the average voltage increment δV to the average current density i_{ave}. As the particle approaches the electrode, δR decreases and converges to a non-zero value. When the entrapment process is complete, the resistance variation will vanish due to no further change of voltage.

The effect of fluid motion in the particle incorporation process will be considered in future work. The lateral migration of the fluid will be considered which will change the concentration field, which in turn will cause the current density to change.

References

1 J.L. Stojak and J.B. Talbot, *J. Electrochem. Soc.*, **146**, 4504 (1999).
2. J.L. Stojak, J. Fransaer, and J.B. Talbot., *Advances in Electrochemical Science and Engineering*, **7**, 193 (2002).
3. J.O. Dukovic, *IBM J. Res. Develop.*, **34**, 693 (1990).
4. E.C. Hume III., W.M. Deen, and R.A. Brown, *J. Electrochem. Soc.*, **131**, 1251 (2001).
5. S. Osher and R. Fedkiw, *Level Set Methods and Dynamic Implicit Surfaces*, Springer-Verlag, New York (2003).
6. R.E. Bank, PLTMG: A Software Package for Solving Elliptic Partial Differential Equations. User's Guide 9.0. Department of Mathematics, University of California at San Diego, (2004).

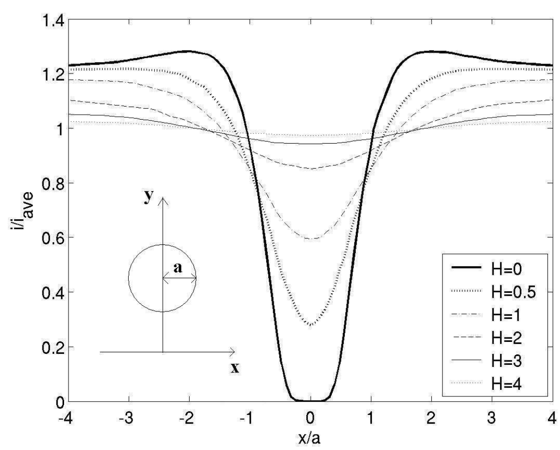

Figure 1. The normalized current density distribution vs. the normalized distance (H=y/a) of a particle for a primary current distribution. Here i_{ave} is the normalized average current density by $RT\kappa/Fa$(mA/cm^2), where κ is the conductivity ($\Omega^{-1}cm^{-1}$).

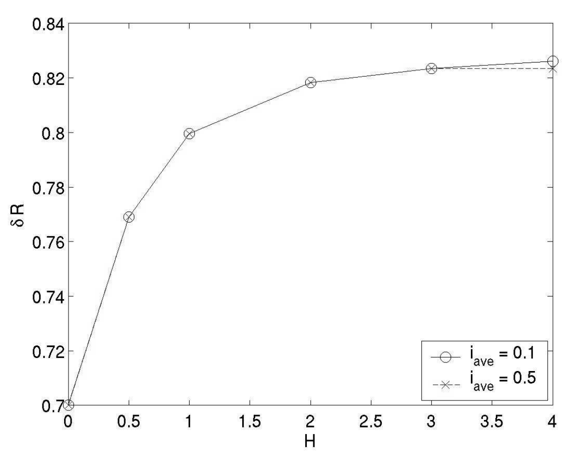

Figure 2. The resistance variation δR vs. H for primary current distribution. Here the voltage is normalized by RT/F and δR (=$\delta V/i_{ave}$) is non-dimensionalized by $a/\kappa(\Omega)$.