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Linear instability of the electroforming process

A.A. LACEY,[1] J.A. McGEOUGH[2] and M. SHILLOR[3]
[1]*Department of Mathematics, Heriot-Watt University, Riccarton, Edinburgh EH14 4AS, UK;*
[2]*Department of Mechanical Engineering, University of Edinburgh, Edinburgh, UK;*
[3]*Department of Mathematics, Imperial College, London, UK*

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Abstract. In a mathematical model for the electroforming process a linear stability analysis shows that the metal/electrolyte interface is unstable under small perturbations. Thus the model is ill-posed, so a different dynamic condition on the free boundary is suggested which allows nonuniformities to form but eliminates infinite front velocities, thus reflecting better the actual process.

1. Introduction

In electroforming a metal layer is electrodeposited upon the cathode, or mandrel, of an electrolytic cell. A major industrial problem associated with the general electroforming process is the production of a uniform thickness of metal when the mandrel has a general (irregular) shape. The formation of a nonuniform thickness is often a source of mechanical weakness with many electroformed components. In practice this difficulty is sometimes overcome by the periodic reversal of the electrode polarity. The electrodeposition takes place over a period of time that is followed by a period of dissolution, controlled such that a uniform thickness for the electroformed component is obtained.

A mathematical model for this process was considered in [1] where some of its properties are discussed and results of some numerical solutions are presented. Following industrial practice the model was considered not only for electrodeposition but also for dissolution.

The purpose of this present note is to perform a Mullins-Sekerka type of linear stability analysis (see e.g. [2]) on a generalized version of the model [1] and to show that the electroforming model has an unstable interface. This means that small perturbations that appear on the forming surface (the free surface) grow exponentially with time. Moreover as the wavelength of the perturbation becomes smaller the growth-rate becomes quicker. Therefore in the absence of any stabilizing mechanism in the model, the velocity of the free boundary becomes infinite and so the model breaks down. On the other hand, it follows from the stability analysis that the model for the dissolution process has a linearly stable interface, and small perturbations decay exponentially. But this is exactly the model for the electrochemical machining process that is known to be stable (see e.g. [3]). These two results concerning the mathematical models seem to correspond to the events actually observed in the industrial process. A similar behaviour can be found in the Hele-Shaw problem (see e.g. [3]) both experimentally and in the mathematical model. The filling of the cell with a viscous fluid is stable while the sucking of the fluid is unstable. In the model the free boundary develops a cusp and so the velocity becomes infinite. In practice "fingers" can be seen to form.

The generalization of the model in [1] consists of the replacement of the standard dynamic condition by a more general one that can take into account current cut-offs. In the former the velocity of the free boundary (i.e. the rate of growth of the interface) is taken to be proportional to the normal derivative of the electric potential (the current) on the free boundary. Our generalization is in taking the velocity to be a nonlinear function of the current provided that the current is above some threshold level, otherwise the interface remains stationary. Such a type of relationship between the growth rate and the current was proposed in [4] in a model for the electrodischarge machining process.

Finally we propose a different dynamic condition for the deposition rate that takes into account the overpotentials that are likely to exist on the free boundary. This condition prohibits infinite velocities and thus partially stabilizes the model but can still lead to the formation of nonuniformities, so possibly giving a better correspondence between the model and the physical process.

The generalized model is given in Section 2 and the linear stability analysis is performed in Section 3.

2. The mathematical model

We consider a generalized model for the electroforming process. A region filled with electrolyte has two electrodes on its boundary. One of the electrodes is connected to a source of positive constant potential while the other, the mandrel, is connected to zero potential. As a consequence of the flow of current through the electrolyte metal deposition takes place on the mandrel. Thus the forming surface (the free boundary) moves into the region. We neglect any chemical reactions in the electrolyte and the effect of any fluid flow. Also for simplicity we consider just two-dimensional problems and use nondimensional variables. We let $\phi = \phi(x, y, t)$ be the electric potential in $\Omega = \{(x, y)\colon -\infty < x < \infty, Y(x, t) < y < 1\}$, the region containing the solution at time $t \geqslant 0$. This geometry is taken as in [1] for the sake of simplicity, see Fig. 1.

Charge conservation implies $\nabla^2 \phi = 0$ in Ω, all $t \geqslant 0$. On the electrode $\Gamma_1 = \{(x, y)\colon -\infty < x < \infty, y = 1\}$ we apply $\phi = 1$. Now let $\Gamma = \{(x, y)\colon -\infty < x < \infty, y = Y(x, t)\}$

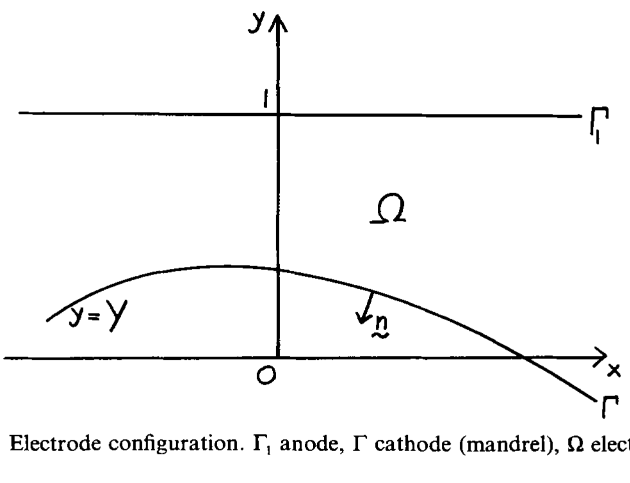

Fig. 1. Electrode configuration. Γ_1 anode, Γ cathode (mandrel), Ω electrolyte.

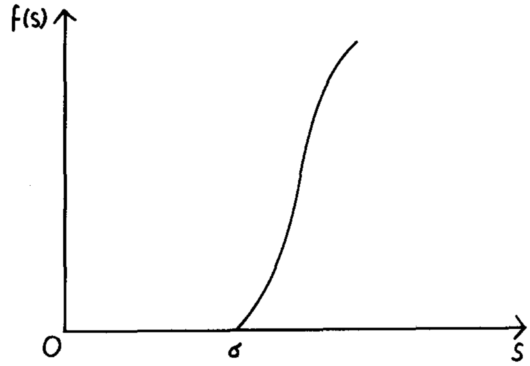

Fig. 2. The form of $f(s)$. σ is the cut-off current.

be the free boundary at time t. Then on this boundary we have to specify two conditions. The first is $\phi = 0$, the second, sometimes called a dynamic condition, is usually taken as outward normal velocity of Γ, $v = \phi_n$, where **n** is the unit vector to Γ out of Ω and $\phi_n = \partial\phi/\partial n$ is the normal derivative. Thus the rate of deposition $(-v)$ is assumed to be proportional to the current density (ϕ_n). Instead we use a more general condition $v = -f(|\phi_n|)$ where $f = f(s)$ can be a fairly general function that satisfies the following conditions: it is Lipschitz continuous in $0 \leqslant s < \infty$, i.e. there is some constant K such that $|f(s_1) - f(s_2)| \leqslant K|s_1 - s_2|$ for $0 \leqslant s_1, s_2 < \infty$; $f(s) \to \infty$ as $s \to \infty$; there exists a cut-off current $\sigma \geqslant 0$ such that $f(s) = 0$ for $0 \leqslant s \leqslant \sigma$; $f(s) > 0, f'(s) > 0$, and f is continuously differentiable for $s > \sigma$, see Fig. 2.

Therefore the mathematical model for the process is

$$\nabla^2\phi = 0 \quad \text{in} \quad \Omega, \tag{1}$$

$$\phi = 1 \quad \text{on} \quad \Gamma_1, \tag{2}$$

$$\phi = 0 \quad \text{on} \quad \Gamma, \tag{3}$$

$$v = -f(|\phi_n|) \quad \text{on} \quad \Gamma. \tag{4}$$

Note that a model for the process of dissolution (electromachining) is obtained from (1)–(4) by changing the sign in (4), i.e.

$$v = f(|\phi_n|). \tag{4'}$$

3. Linear stability analysis

We perform a Mullins-Sekerka type linear stability analysis (see e.g. [2]) on the interface Γ in the model (1)–(4). We assume that a small perturbation appears at some time, which we take to be at $t = 0$, on a flat interface, taken as $y = 1 - 1/A$ (at $t = 0$), and show that its amplitude has an exponential growth rate. Then the unperturbed, hence independent of x, absolute value of the normal derivative of ϕ on Γ at $t = 0$ is A. If $A < \sigma$ then by (4) nothing happens (no deposition) provided the perturbation of $-\phi_n$ is smaller than $\sigma - A$. So let $A > \sigma$ and let $U = -f(A)$ be the unperturbed front velocity (U is the outward velocity, i.e. in the opposite direction to growth and hence negative). Let

$$Y(x, t) = 1 - 1/A - Ut + \varepsilon a(t) \sin kx + \dots, \tag{5}$$

the perturbation being small initially, $\varepsilon \ll 1$ and $0 < a(0) = O(1)$, $k > 0$ is the wave number ($2\pi \times$ the reciprocal of the wavelength) of the perturbation. The perturbed potential, that has to satisfy (1), is

$$\phi = A(y - 1 + 1/A + Ut) + \varepsilon b(t) \exp(-k(y - 1 + 1/A + Ut)) \sin kx + \dots . \tag{6}$$

(We actually replace the earlier region Ω with $Y < y < \infty$ and require that the perturbation decays to 0 as $y \to \infty$, or equivalently keep condition (2) but suppose that $k \gg 1$ which gives (6) on neglecting exponentially small terms.)

In order to satisfy (3) on $y = Y$ we have from (5) and (6) that

$$A(1 - 1/A - Ut + \varepsilon a \sin kx + \cdots - 1 + 1/A + Ut) + \varepsilon b \sin kx + \cdots = 0,$$

hence to order ε

$$b(t) = -Aa(t). \tag{7}$$

Now we want (4) to be satisfied. First note that

$$v = -\partial Y/\partial t + O(\varepsilon^2) = U - \varepsilon \dot{a} \sin kx + \dots \tag{8}$$

where $\dot{a} = \mathrm{d}a/\mathrm{d}t$. Next notice that $\partial/\partial n = -\partial/\partial y + \varepsilon \partial Y/\partial x\, \partial/\partial x + \dots$, hence

$$\phi_n = -A + \varepsilon bk \exp(-k(y - 1 + 1/A + Ut)) \sin kx + \dots \tag{9}$$

where $y = Y$ and so $\exp(-K(y - 1 + 1/A + Ut)) = 1 + O(\varepsilon)$. Inserting (8) and (9) into (4) gives, to order ε,

$$U - \varepsilon \dot{a} \sin kx \sim -f(A - \varepsilon bk \sin kx),$$

so

$$-U + \varepsilon a \sin kx \sim f(A) - f'(A)\, \varepsilon bk \sin kx,$$

but $U = -f(A)$, hence $\dot{a} \sim -kbf'(A)$, and from (7) it follows that

$$\dot{a}(t) \sim Akf'(A)\, a(t). \tag{10}$$

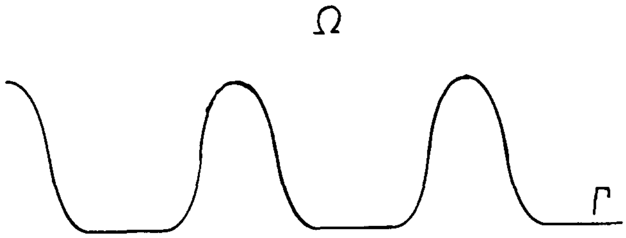

Fig. 3. Growing fingers for the special case $A = \sigma$.

Now $f'(A) > 0$ by our assumptions, hence we obtain from (10)

$$a(t) \sim B \exp(ct) \tag{11}$$

for appropriate $B > 0$ and $c > 0$. Thus the amplitude of the perturbation grows faster for larger wave number k (smaller wavelength). It may be conjectured (see e.g. [5]) that almost immediately a cusp forms in the free boundary and the velocity there is infinite.

The special case of $A = \sigma$ is rather more complex. In this case the periodic perturbation of the only just stationary free boundary is no longer sinusoidal but instead consists of straight stationary parts and exponentially growing fingers (see Fig. 3).

On the other hand in the model for dissolution (4) is replaced by (4′), i.e. $v = f(|\phi_n|)$, and repeating the calculations above now gives

$$\dot{a} \sim -Akf'(A)a \tag{12}$$

instead of (10), and since the right-hand side in (12) is negative it leads to exponential decay of the perturbation, i.e. linear stability. Clearly all these results apply to the special cases of the usual models, i.e. $\sigma = 0$ and $f(s) = s$ so that (4) is $v = \phi_n$ and (4′) is $v = -\phi_n$.

4. Conclusion

We have found that the generalized model for the electroforming process has a linearly unstable free boundary. Thus mathematically the model is likely to be ill-posed as generally solutions are unlikely to exist. This implies that numerical solutions to this model should also fail to exist or, if a crude enough mesh is used, to misbehave as a refined mesh is introduced. On the other hand in the real process, although nonuniformities do occur, the velocity of the free boundary remains bounded. This suggests that the condition (3) in the model should be replaced by an overpotential relation (see [6]) between the current and the potential:

$$\phi_n = -g(\phi) \tag{13}$$

where $g(s)$ is a positive function for $s > 0$ and Lipschitz continuous with $g(0) = 0$ and $g'(s) > 0$ for $s \geqslant 0$, see Fig. 4.

The stability now follows from the observation that since $0 \leqslant \phi \leqslant 1$ ($\phi = 1$ on Γ_1) then $\max(-v) = \max f(|\phi_n|) \leqslant f(g^{-1}(1))$ and hence the velocity of the free boundary is bounded. Note, however, that nonuniformities can still grow.

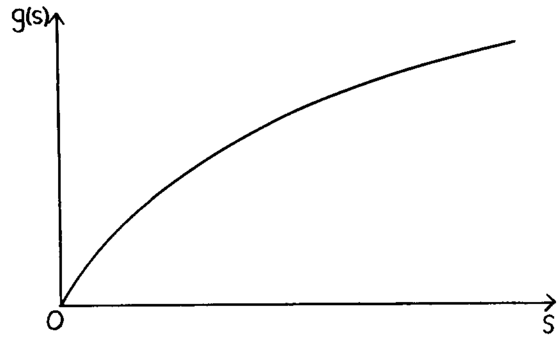

Fig. 4. The overpotential $g(s)$.

We also remark that mathematical models with such boundary conditions have been considered in one space dimension for the heat equation, see e.g. [7] and references there.

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