

# Surface Boundary Conditions for Small Amplitude Waves on a Falling Liquid Film

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Several years ago the author (1964) analyzed the stability characteristics of falling liquid films in terms of the Orr-Sommerfeld equation and appropriate boundary conditions for the gas-liquid interface. In the absence of surfactants, the analysis provided excellent agreement with the experiments of Jones and Whitaker (1966), and further theoretical and experimental studies were performed (Strobel and Whitaker, 1969; Whitaker and Jones, 1966) on the basis of the original work. In a more recent paper, Lin (1970) has characterized his study of this problem by the statement that "the present analysis differs from the previous works in the treatment of the surface boundary conditions." Regarding the work of Whitaker and Jones (1966), he goes on to say that "the free surface boundary conditions are not exact" and then offers an explanation for the "apparent agreement between their results and Benjamin's results."

The purpose of this note is simply to point out that the free surface boundary conditions used by Whitaker (1964) and later by Whitaker and Jones (1966) are identical to those used by Lin, thus the explanation offered by Lin is of no consequence and should be disregarded.

The comparison between the two works is best seen by examining the tangential stress condition for a surface exhibiting a constant surface stress or surface tension. Under these circumstances the tangential stress condition of Whitaker (1964) reduces to

$$F'' + \alpha^2 F = 2, \quad Y = 0 \quad (1)$$

where the primes denote differentiation with respect to  $Y$ , and the kinematic surface condition is

$$F = C - 1, \quad Y = 0 \quad (2)$$

Here Equations (1) and (2) are identical to Equations (16) and (10) respectively in the original paper by Whitaker (1964), and Equations (17) and (12) in the paper by Whitaker and Jones (1966) provided one takes the surface stress to be constant. The symbols  $F$  and  $C$  represent the dimensionless disturbance stream function and dimensionless complex growth rate constant respectively. Noting that the kinematic condition can be expressed as

$$\frac{F(0)}{C - 1} = 1 \quad (3)$$

we can multiply the righthand side of Equation (1) by one to obtain

$$F''(0) + \alpha^2 F(0) = 2 \left[ \frac{F(0)}{C - 1} \right] \quad (4)$$

This can be rearranged to yield

$$F''(0) + \left( \alpha^2 - \frac{2}{C - 1} \right) F(0) = 0 \quad (5)$$

In Lin's formulation of the stability problem the stream function and growth rate constant are made dimensionless by use of the average velocity, as opposed to the surface velocity, thus the following relations exist:

$$F = \left( \frac{2}{3} \right) \phi \quad (6)$$

$$C = \left( \frac{2}{3} \right) c \quad (7)$$

Here  $\phi$  and  $c$  are Lin's dimensionless stream function and growth rate constant respectively. Substitution of Equations (6) and (7) into Equation (5) leads to

$$\phi''(0) + \left( \alpha^2 - \frac{3}{c - 3/2} \right) \phi(0) = 0 \quad (8)$$

If one defines  $c' = c - 3/2$  we obtain

$$\phi''(0) + (\alpha^2 - 3/c') \phi(0) = 0 \quad (9)$$

which is Lin's Equation (8) for the tangential stress condition when the surface stress is constant. Similar algebraic manipulations can be used to show that the normal stress condition used by Lin is identical to that given earlier by Whitaker and Jones (1966). In addition, the work of Benjamin (1957, 1964), Anshus and Goren (1966), and of Anshus and Acrivos (1967), should be noted as having these universally accepted forms for the tangential and normal stress conditions.

It should be clear at this point that the formulation of the surface boundary conditions given by Lin is identical to that given by Whitaker and Jones. Further study indicates that the entire formulation of the stability problem given by Lin is identical to the earlier work of Whitaker and Jones. For example, the result listed after Lin's Equation (47) for the critical Reynolds number is identical to Equation (42) of Whitaker and Jones, and is precisely the result obtained in 1964 by direct numerical integration of the Orr-Sommerfeld equation for a vertical falling liquid film. This result for the critical Reynolds number associated with insoluble surfactants has also been presented by Benjamin (1964) and by Anshus and Acrivos (1967). It was used originally by Strobel and Whitaker (1969) to estimate surface elasticities from wave inception line data, and more recently by Cerro and Whitaker (1971).

Regarding soluble surfactants, Lin comments that: "No direct comparison between the present results and the results of Whitaker and Jones is possible, because their results for the case of soluble surface-active agents were obtained as a fourth-order perturbation solution to an eigenvalue problem in which two of the boundary conditions are correct only up to the first-order approximation." However, we have seen that the difference referred to by Lin is nonexistent. A possible source of discrepancy for

the soluble surfactant case is that Lin apparently expanded the surface concentration gradient in terms of  $\alpha$ , whereas the work of Whitaker and Jones made a rather careful point of noting that the soluble surfactant case had to be treated in terms of an expansion in the square root of  $\alpha$ . This becomes clear if one notes that the expression for the surface concentration gradient for a soluble surfactant contains the square root of  $\alpha$  in the denominator.\* Under these circumstances we are dealing with a function of the form,

$$\psi = \frac{A}{B + \sqrt{\alpha}} \quad (10)$$

and a Taylor series expansion in terms of  $\alpha$  leads to

$$\psi(\alpha) = \psi|_{\alpha=0} + \alpha \left( \frac{\partial \psi}{\partial \alpha} \right)_{\alpha=0} + \frac{\alpha^2}{2} \left( \frac{\partial^2 \psi}{\partial \alpha^2} \right)_{\alpha=0} + \dots \quad (11)$$

One quickly sees that

$$\left( \frac{\partial \psi}{\partial \alpha} \right)_{\alpha=0} = \left( \frac{\partial^2 \psi}{\partial \alpha^2} \right)_{\alpha=0} = \left( \frac{\partial^n \psi}{\partial \alpha^n} \right)_{\alpha=0} = \infty \quad (12)$$

thus  $\psi$  must be expanded in terms of  $\sqrt{\alpha}$  not  $\alpha$ . Lin apparently circumvented this difficulty by expanding the numerator of the term representing the surface concentration gradients, but not the denominator. Under these

circumstances it is not surprising that the results of Lin are different from those of Whitaker and Jones.

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\* See, for example, the equation for  $\Sigma'$  following Lin's Equation (13).

## Comments on Surface Boundary Conditions for Small Amplitude Waves on a Falling Liquid Film

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The treatment of the boundary conditions for the problem of the stability of a contaminated liquid film by Lin (1970) is different from that by Whitaker and Jones (1966). Aside from the fundamental differences in the modeling of the monolayer, the following differences in the treatment of the boundary conditions are important. The problem of the present interest, in terms of mathematics, is a linear homogeneous eigenvalue problem. Hence, the eigenfunction can be obtained only up to an arbitrary constant multiplier. While the normalization of the eigenfunction is effected by Lin with  $F(0) = 1$ , the normalization by Whitaker and Jones is based on  $F(0) = C - 1$ . The latter normalization has one disadvantage in

that it depends on the unknown eigenvalue  $C$ . Therefore, the normalization must be effected in each step of the perturbation solution. Consequently, the eigenvalue must be obtained in each step from five simultaneous algebraic equations instead of from four equations, which is the case in Lin's treatment. Moreover, the term  $i\alpha^3 N_{Re} N_{We}$  in Equation (16) of Whitaker and Jones is treated as a term of  $O(\alpha^3)$ . Therefore, this term is not retained in their first three order perturbations. As is pointed out by Krantz and Goren (1970), this surface tension term is of order  $\alpha$  according to the laboratory observations and must be included in the solution. As a direct consequence of this omission,  $C_1$  given in (36) of Whitaker and Jones differs