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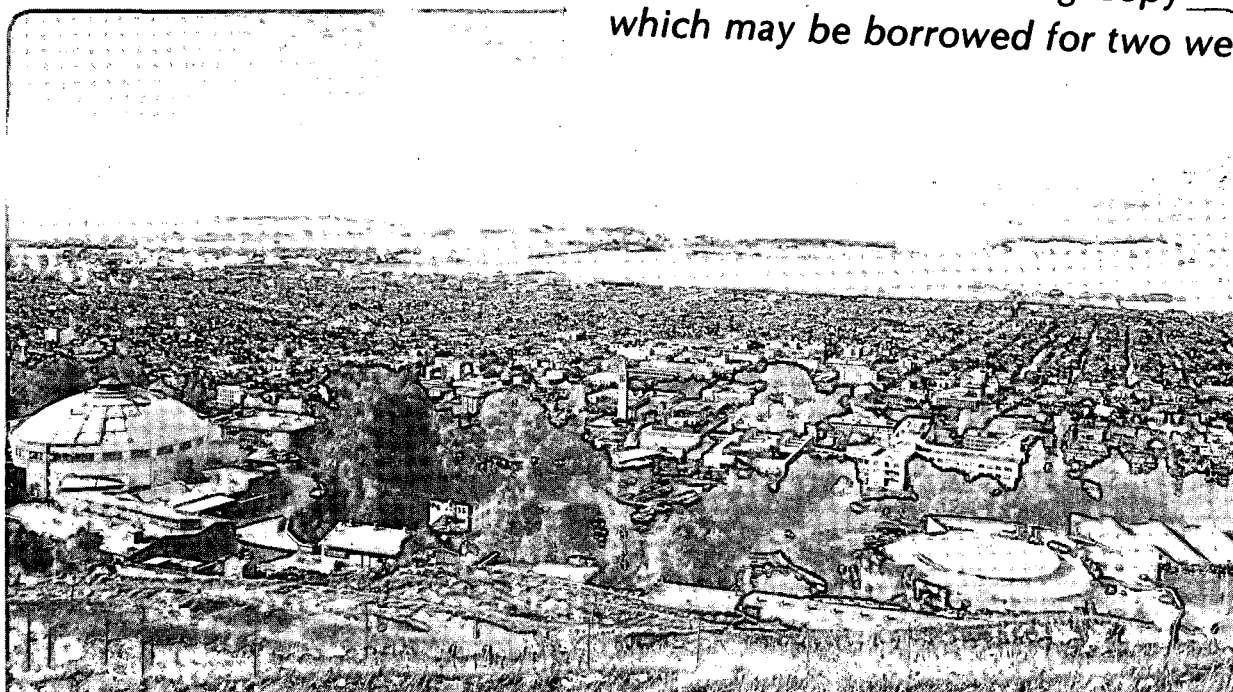
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The Influence of Surfactants on the Flow of Long Bubbles through
Cylindrical Capillaries

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THE INFLUENCE OF SURFACTANTS ON THE FLOW OF LONG BUBBLES THROUGH CYLINDRICAL CAPILLARIES

By

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INTRODUCTION

Foam is a promising fluid for increasing mobility control in underground enhanced oil recovery. An understanding of the fundamentals of foam flow in porous media may be gained by studying the flow of surfactant-stabilized bubble trains in capillary tubes (1,2). Thus, by scaling arguments similar to those used for non-Newtonian polymer solutions (3), it appears possible to predict foam-flow behavior in porous media using measurements of and a theory for bubble flow in tubes (4).

This work considers the behavior of a single gas bubble in a capillary with the continuous liquid phase containing surfactant whose concentration is near or above the critical micelle concentration. We extend Bretherton's analysis for a clean gas bubble (5) to include the effects of a kinetically hindered, soluble surfactant in the liquid phase on the shape of the bubble, and on the resulting pressure drop across the bubble.

KINEMATICS

A schematic of a long bubble flowing through a tube filled with surfactant solution is shown in Figure 1. The reference frame is such that the bubble is stationary with the walls moving past at a velocity of $-U$.

The height of the bubble interface, h , is measured from the tube wall. In this analysis, the undistorted bubble radius is always greater than the tube radius, and the bubble is longer than twice the tube radius. Consequently, there is a region of constant liquid film thickness, h_0 , in the middle of the bubble.

The shape of the front and rear menisci change as a result of the resistance to bubble flow. This deviation in bubble shape establishes the dynamic pressure drop across the bubble.

The expected surfactant distribution is also shown in Figure 1. Recirculation eddies in the liquid phase lead to stagnation perimeters around the bubble, as shown by the heavy black dots on the interface. Near the bubble front, surfactant molecules are swept away from the stagnation perimeter and are not instantaneously replenished from the bulk solution. Accordingly, a surface stress develops along the interface from low to high surface tension (i.e., high to low surfactant concentration). Surface stresses at the rear stagnation perimeter also occur. There are no surface velocity gradients in the thin film portion of the bubble, and the surface tension returns to its equilibrium value. Constant surface tension in the thin film portion of the bubble allows the bubble to be treated as if it were infinite in length and thus, the front and back menisci may be treated separately.

In examining either end of the bubble, the interface can be divided into three distinct regions, as shown in Figure 1. Region I is characterized by a constant thin film thickness, region III is of constant curvature where the shape of the bubble can be described by a sphere, and region II is a transition region between the two asymptotic limits. For

small capillary numbers, $Ca = \mu U / \sigma_0$, surface tension forces dominate and region II conforms closely to the capillary wall so that the lubrication approximation may be used throughout.

EQUATION DEVELOPMENT

The normal stress balance is used with the lubrication approximation to obtain the pressure at any point in the liquid phase as a function of the position of the bubble interface, h . The liquid velocity is determined by using the Navier-Stokes equation with the following boundary conditions. The liquid velocity at the tube wall equals $-U$ and the surface stress at the gas-liquid interface is given by the gradient in the surface tension, $-d\sigma/dx$. A macroscopic continuity balance states that the average flow rate at any x position along the bubble must equal the flow rate in the thin film region. Combination of the normal stress balance, the Navier-Stokes equation, and the macroscopic continuity balance results in a third order differential equation for the position of the bubble interface as a function of x .

A second equation is needed to determine the surface tension as a function of axial position. This expression is provided by the surface species continuity balance. A kinetic rate limitation is assumed for surfactant concentrations above the critical micelle concentration. The surface species continuity balance requires that the flux of surfactant along the bubble interface must be balanced by the net rate of adsorption and desorption of surfactant from the liquid solution. This balance gives the surface surfactant concentration, Γ , as a function of position, x . The quasistatic adsorption assumption then allows calculation of the surface tension as a function of position along the bubble interface.

The equations for the position of the bubble interface and the surfactant distribution are nondimensionalized by the Bretherton scaling (5) which, after eliminating terms of higher order in capillary number, yield the following:

$$\eta^3 \eta_{\xi\xi\xi} - \frac{3}{2} \alpha \eta^2 \theta_{\xi} - (\eta - \eta_0) = 0, \quad 1)$$

and

$$\frac{1}{\beta} \left[3\eta \eta_{\xi} \eta_{\xi\xi\xi} + \frac{3}{2} \eta^2 \eta_{\xi\xi\xi\xi} - 3\alpha \eta_{\xi} \theta_{\xi} - 3\alpha \eta \theta_{\xi\xi} \right] = -\theta, \quad 2)$$

where η is the dimensionless height of the bubble interface, η_0 is the dimensionless thin film thickness, ξ is the dimensionless x distance, and θ is the dimensionless surface surfactant concentration such that when $\theta=0$, the surfactant concentration is at its equilibrium value, Γ_0 . The subscript, ξ , refers to differentiation with respect to ξ . α is an equilibrium dimensionless parameter which measures how strongly the surface tension depends on the surfactant distribution and corresponds to the Gibbs elasticity. β is a second dimensionless parameter which reflects the kinetics of the adsorption process. $\alpha=0$ and $\beta=\infty$ reduce Equations 1) and 2) to the case of constant surface tension solved by Bretherton. The ratio α/β is the elasticity number, characterizing the stabilizing effect which the presence of adsorbed surfactant molecules have on the bubble interface.

PERTURBATION SOLUTION

The equations are solved for a small deviation from the constant tension bubble case. This is done by a regular perturbation expansion in $1/\beta$ about the constant tension solution. The resulting equations to zero and first order in $1/\beta$ are:

0[1]

$$\theta^0 = 0 \quad , \quad 3)$$

$$(\eta^0)^3 \eta_{\xi\xi\xi}^0 - \eta^0 + 1 = 0 \quad , \quad 4)$$

0[1]
 β

$$\theta^1 = -\frac{3}{2} \eta_{\xi}^0 \frac{1}{(\eta^0)^2} \quad , \quad 5)$$

and

$$\begin{aligned} & (\eta^0)^3 \eta_{\xi\xi\xi}^1 + 3(\eta^0)^2 \eta_{\xi\xi\xi}^0 \eta^1 - \eta^1 \\ & - \frac{3}{2} \frac{1}{\eta^0} \left[3(\eta_{\xi}^0)^2 - \frac{3}{2} \eta^0 \eta_{\xi\xi}^0 \right] = \eta_o^1 \quad , \quad 6) \end{aligned}$$

where the superscripted variables refer to the order in $1/\beta$. Equations 3) and 4) represent the constant tension equations with an equilibrium surfactant distribution, and have been solved by Bretherton. Equations 5) and 6) can be solved to obtain the first order contribution to the surface position and surfactant concentration due to the presence of a surfactant. Details of the numerical calculations are given by Ginley (6).

The first order contribution to the surfactant concentration distribution in Equation 5) can be determined directly from the solution for η^0 . Equation 6) has three homogeneous solutions and two particular solutions. The overall solution was obtained by the method of matched asymptotic expansions after performing three separate numerical integrations (6)

RESULTS

The numerical solutions for the front of the bubble are shown in Figure 2. The total dimensionless film thickness, η , is shown as a function of a dimensionless axial position ζ where ζ differs from the coordinate ξ by an additive constant. Recall that $\eta = \eta^0 + (1/\beta)\eta^1$. The

Bretherton solution is that for constant surface tension, $\alpha/\beta=0$, and the second curve is shown for a value of the elasticity number, α/β , equal to one. Note that the only significant deviations in the bubble profile occur in the region between $\zeta=0$ and $\zeta=5$ which is inside of the transition region (region II in Figure 1). Viscous stresses are important in this transition region and the surface tension gradients have a noticeable effect on the shape of the bubble interface. Figure 3 shows total profiles for the bubble back for various values of α/β . The magnitude and frequency of the oscillations in film thickness become greater with the addition of a surfactant.

The thin film thickness and the pressure drop across the bubble are determined by matching the numerical solution in the inner region II with the outer solution in region III. These are the main results of our analysis:

$$\frac{h^0}{R_T} = 1.337[1 - 1.004 \times 10^{-2} \frac{\alpha}{\beta}] Ca^{2/3} \quad 7)$$

and

$$\frac{\Delta P_{RT}}{2\sigma_o} = 9.40[1 + 0.469 \frac{\alpha}{\beta}] Ca^{2/3} \quad 8)$$

The first term in both Equations 7) and 8) is the constant surface tension contribution and the second term gives the first order contribution resulting from the presence of a soluble surfactant with finite sorption kinetics. Because of the perturbation approach, we find a linear dependence on the surfactant elasticity number. The thin film thickness deviates by only one percent from the constant tension solution when $\alpha/\beta = 1$ whereas the pressure drop across the bubble is significantly greater than the constant

tension value when $\alpha/\beta=1$. Ginley (6) presents a discussion of α and β values and also compares the proposed theory to existing experimental results.

CONCLUSIONS

A regular perturbation expansion in large adsorption rates has been performed about the low capillary number, singular perturbation theory of Bretherton. The pressure drop across the bubble is found to increase upon the addition of surfactant, whereas the thin film thickness decreases slightly. Both the pressure drop and the thin film thickness retain their $2/3$ power dependence on the capillary number found by Bretherton for surfactant-free bubbles.

ACKNOWLEDGMENTS

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NOMENCLATURE

- $Ca = \mu U / \sigma_0$, capillary number, ratio of viscous to surface tension forces
- c_0 - bulk concentration of surfactant, mol/m^3
- h - local film thickness measured from the capillary wall, m
- h_0 - constant thin film thickness, m
- k_1 - adsorption rate constant, $\text{m}^3/\text{mol}\cdot\text{s}$
- P - pressure, Pa
- R_T - capillary tube inner radius, m
- U - bubble velocity, m/s
- x - axial position, m

y - vertical coordinate measure from the tube wall, m
 α - $(-d\sigma_o/d\Gamma_o)(\Gamma_o/\sigma_o)$, fractional Gibbs elasticity
 β - $0.643(k_1 c_o \Gamma_{\max}/\Gamma_o)(R_T \mu/\sigma_o)$, Damkohler number
 Γ - surface excess concentration, mol/m^2
 Γ_o - equilibrium surface excess concentration, mol/m^2
 $\Gamma_{\max}$ - surface excess concentration corresponding to monolayer coverage, mol/m^2
 ζ - ξ + constant, dimensionless axial coordinate
 η - h/h_o^0 , dimensionless film thickness
 η_o - h_o/h_o^0 , dimensionless thin film thickness
 θ - $(\Gamma/\Gamma_o - 1)(3Ca)^{-2/3}$, dimensionless relative surface excess concentration
 μ - fluid viscosity, mPa's
 ξ - $x/h_o^0(3Ca)^{-1/3}$, dimensionless axial coordinate
 σ - surface tension, mN/m
 σ_o - equilibrium surface tension, mN/m
 τ_s - tangential stress at the gas/liquid interface, mN/m^2

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FIGURE CAPTIONS

- Figure 1. Flow of a single gas bubble through a liquid-filled cylindrical capillary. The liquid contains a soluble surfactant whose distribution along the bubble interface is sketched.
- Figure 2. The bubble front profile for a surfactant-free interface (Bretherton) and a surfactant-laden interface ($\alpha/\beta=1$)
- Figure 3. The bubble rear profile for a surfactant-free interface (solid line) and a surfactant-laden interface (various dashed lines).

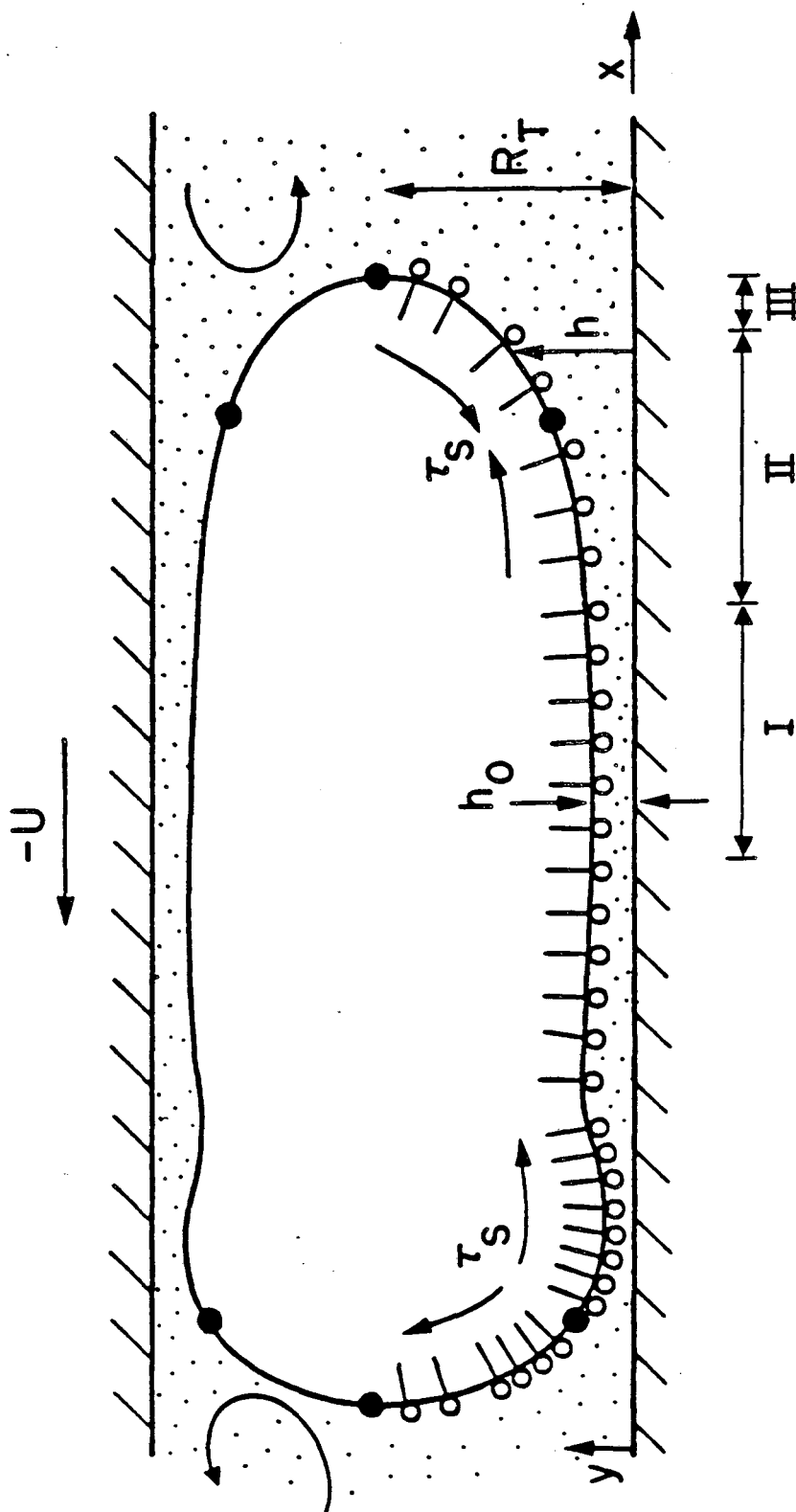


Figure 1

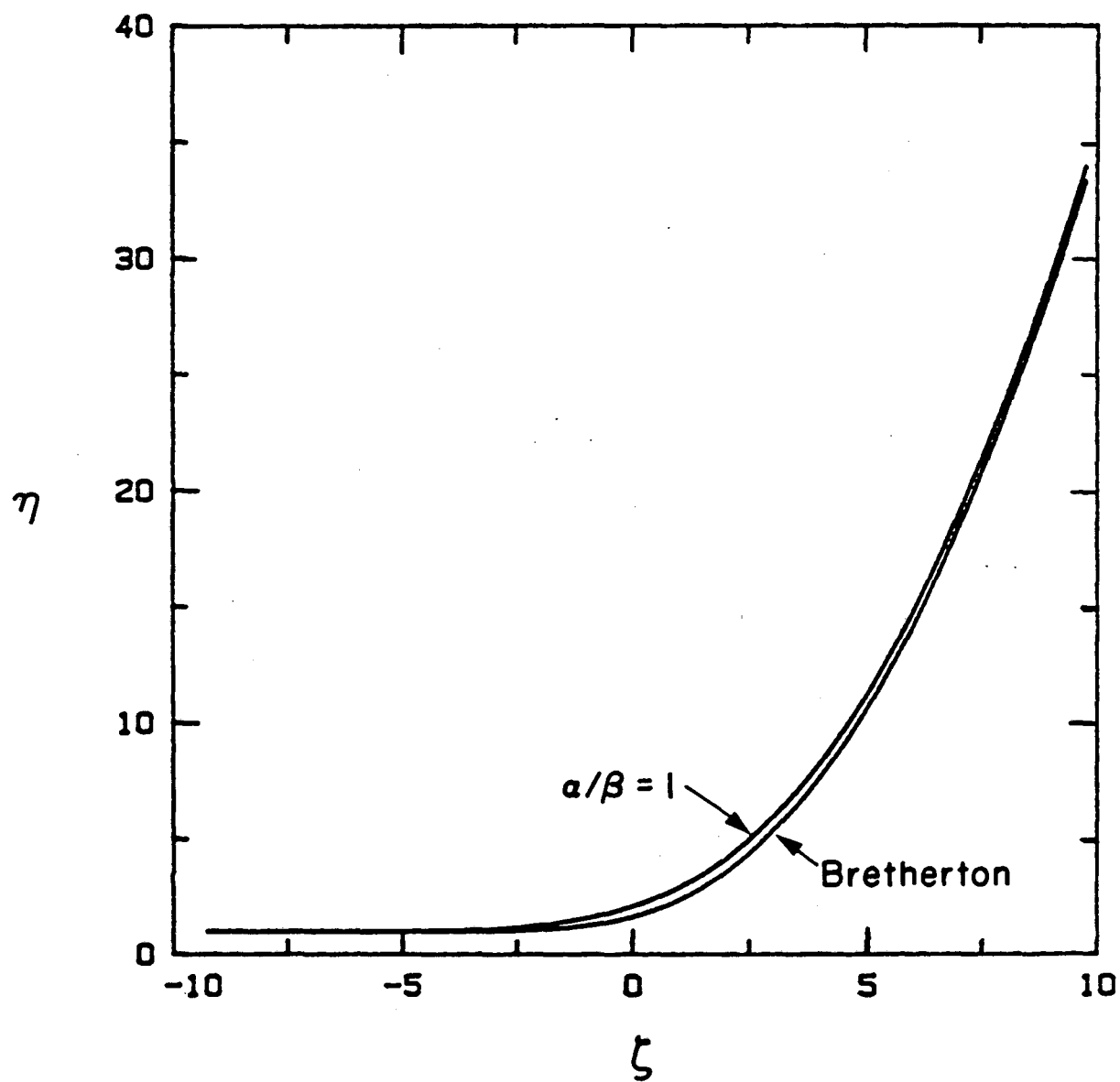


Figure 2

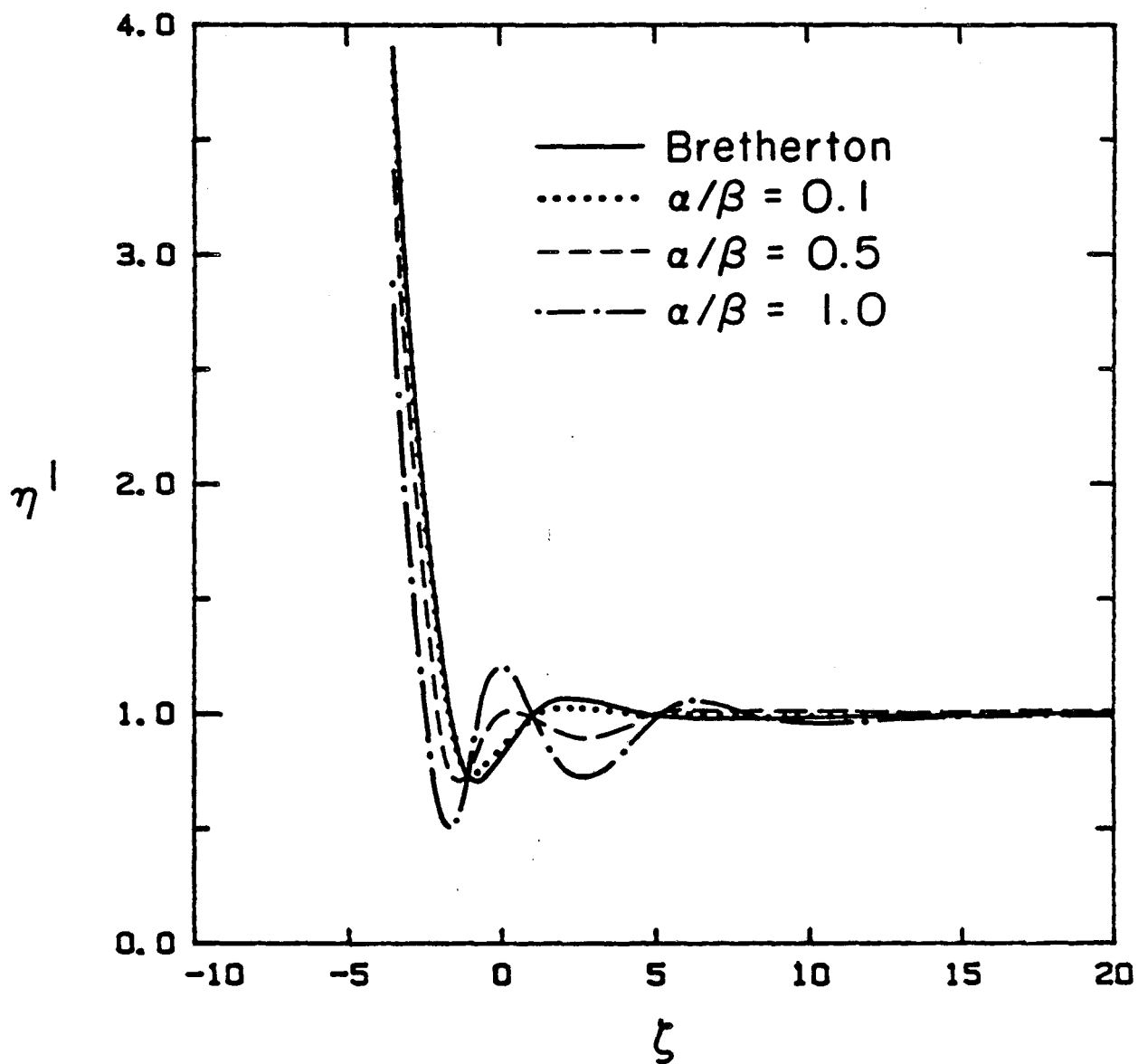


Figure 3

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