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Numerical simulation of coextrusion process of viscoelastic fluids using the open boundary condition method

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Abstract

Numerical simulation of coextrusion process of viscoelastic fluids within a die has been carried out. In the coextrusion process velocity profile at the outflow boundary is not known a priori, which makes it difficult to impose the proper boundary condition at the outflow boundary. This difficulty has been avoided by using the open boundary condition (OBC) method. In this study, elastic viscous stress splitting (EVSS) formulation with streamline upwind (SU) method has been used in the finite element method. In order to test the validity of the OBC method, comparison between the results of fully developed condition at the outlet and those of OBC has been made for a Newtonian fluid. In the case of upper convected Maxwell (UCM) fluid, the effect of outflow boundary condition on the interface position has been investigated by using two meshes having different downstream lengths. In both cases, the results with the OBC method showed reasonable interface shape. In particular, for the UCM fluid the interface shape calculated with OBC was independent of the downstream length, while the results with the zero traction condition showed oscillation of interface position close to the outlet. Viscosity difference was found to be more important than elasticity difference in determining the final interface position. However, the overshoot of interface position near the confluent point increased with elasticity.

Keywords : open boundary condition, coextrusion, upper convected maxwell model, finite element method

1. Introduction

Coextrusion is a process in which two or more polymers are extruded and combined in a feedblock or a die to form a single structure with multiple layers. Coextrusion has been widely used for producing many commercial multilayer plastic products. The unique feature of the coextrusion process is the stratified flow where different materials are separated by an interface. The determination of the interface position of coextrudated polymers is important for obtaining desirable products.

Experimental studies of the coextrusion process of polymer melts have shown that the interface shape between the fluids is dependent upon the viscosity and the elastic properties of polymer melts. In the case of purely viscous fluids, less viscous fluid tends to wrap the fluid that is more viscous. For the viscoelastic fluids, viscosity difference predominates over the elasticity difference in determining the interface shape. However, if the viscosity of two polymers is the same, more elastic component tends to encapsulate less elastic one (Han, 1981; Kahn and Han, 1976; Southern and Ballman, 1975; White *et al.*, 1972).

The finite element method has been widely used for free boundary problems such as die swell or coextrusion due to the ability of handling complex boundaries. In the coextrusion process, pressure and extra stresses are discontinuous along the interface. To treat the discontinuity of pressure and extra stresses across the interface, double-node finite element method has been used (Matsunaga *et al.*, 1998; Mavridis *et al.*, 1987). In double-node method, the primitive variables such as extra stresses and pressure have two values at the same spatial location.

In the numerical simulation of viscoelastic flow within a die, velocity profile should be imposed along the outflow boundary unlike the Newtonian case. Thus, for a single fluid flow, fully developed velocity profile has been imposed customarily at the outflow boundary. If the long domain is used for the numerical simulation, the velocity can be fully developed at the outflow boundary. The outflow boundary condition is matched with the solution calculated in the domain of interest in this case. In nonisothermal flow or coextrusion process, however, the velocity profile at the outflow boundary is not known a priori. Therefore,

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we cannot impose appropriate boundary condition at the outlet, which makes it difficult to impose the boundary condition at the outlet in nonisothermal flow or coextrusion process of viscoelastic fluids.

When there is a swelling part outside of a die, both viscoelastic and purely viscous fluids can be used in the numerical simulation of coextrusion process without difficulty in imposing the outflow boundary condition (Mitsoulis, 1986; Luo and Mitsoulis, 1990). For the flow within a die, however, numerical simulations of coextrusion process (or stratified flow) have been performed mainly using the generalized Newtonian fluids due to this difficulty (Mavridis *et al.*, 1987; Dheur and Crochet, 1987; Khomami, 1990). In the case of viscoelastic fluids, Matsunaga *et al.* (1998) used zero normal and tangential traction condition at the outflow boundary for the simulation of coextrusion process using the White-Metzner model instead of imposing the velocity profile. However, because tractions do not vanish at the outflow boundary in the case of viscoelastic fluid, these boundary conditions are not appropriate from the physical point of view. Dheur and Crochet (1989) simulated the two-layer flow through an abrupt expansion or contraction die using the upper convected Maxwell (UCM) model. Musarra and Keunings (1989) used the Oldloyd-B model in the same geometry and compared simulation results with experimental data. Although they mentioned that fully developed velocity profile for viscoelastic fluid was imposed at the outflow boundary, the detailed information was not referred in their paper. Takase *et al.* (1998) performed three-dimensional coextrusion simulation in a duct using the fully developed boundary condition at the outlet for the Phan Thien-Tanner (PTT) model to show the elastic effect on the encapsulation phenomena.

The difficulty in imposing outflow boundary condition can be avoided using the open boundary condition (OBC) method, in which the boundary conditions are not imposed along the outflow boundary. The open boundary condition method allows the solution, calculated in the upstream, to pass through the outflow boundary without undergoing significant distortion and without influencing the interior solution. In OBC method, line (or surface) integral along the outflow boundary is calculated by unknown outflow nodal values, which are obtained as part of solution.

Open boundary condition method leads to an undetermined problem at the level of partial differential equation. However, it yields a well-defined problem at the discrete level. Griffiths (1997) showed that using finite element method of degree p, open boundary condition method is equivalent to imposing the condition that the (p+1)st derivative of the dependent variable should vanish at a point close to the outflow in the one-dimensional advection-diffusion problem. Renardy (1997) also analyzed the errors in using the open boundary condition method for the one-dimensional advection-diffusion problem and showed that the open boundary condition method actually imposes an effective boundary condition.

Open boundary condition method has been applied in various problems. Papanastasiou *et al.* (1992) applied it to the backward facing step problem at high Reynolds number. Sani and Gresho (1994) compared the results obtained in various test problems, which include steady or time-dependent flows in backward facing step geometry and channel for Newtonian fluid. Papanastasiou *et al.* (1996) used the open boundary condition method in the fiber spinning of the viscoelastic fluids to impose the stress at the inlet boundary and verified the admissibility and accuracy of the open boundary condition method. Park and Lee (1999) simulated nonisothermal viscoelastic flow in the contraction die using the UCM fluid. In their study, the results obtained in the meshes having different downstream length were compared to verify the open boundary condition method, and the viscous heating effect on the stress and velocity profiles was also investigated.

In this work, numerical simulation of coextrusion process for viscoelastic fluid within a die is performed using the UCM fluid. To test the validity of the open boundary condition method, the results calculated using the OBC are compared with those obtained using the fully developed boundary condition for a Newtonian fluid. The effect of elasticity and viscosity ratio on the interface position is studied, and the influence of boundary conditions on the solution is also examined.

2. Governing equations and numerical method

The steady state flow of an incompressible viscoelastic fluid is governed by a set of conservation and constitutive equations. The mass and momentum conservation equations are as follows:

$$\nabla \cdot \boldsymbol{v} = 0 \tag{1}$$

$$-\nabla p + \nabla \cdot \boldsymbol{\tau} = 0 \tag{2}$$

where $\boldsymbol{v}$ is the velocity, p is the pressure, and $\boldsymbol{\tau}$ is the extra stress. In this study, the upper convected Maxwell (UCM) model is used for the viscoelastic constitutive equation, and is written as

$$\boldsymbol{\tau} + \lambda \overset{\nabla}{\boldsymbol{\tau}} = \eta(\nabla \boldsymbol{v} + \nabla \boldsymbol{v}^T) \tag{3}$$

where λ is the relaxation time and η is the viscosity. The triangular superscript stands for upper convected derivative, which is defined as:

$$\overset{\nabla}{\boldsymbol{\tau}} = \frac{\partial \boldsymbol{\tau}}{\partial t} + \boldsymbol{v} \cdot \nabla \boldsymbol{\tau} - \nabla \boldsymbol{v}^T \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \nabla \boldsymbol{v} \tag{4}$$

In a coextrusion process, two polymer melts form a stratified flow. The schematic diagram of stratified flow is depicted in Fig. 1. Referring to a local coordinate system,

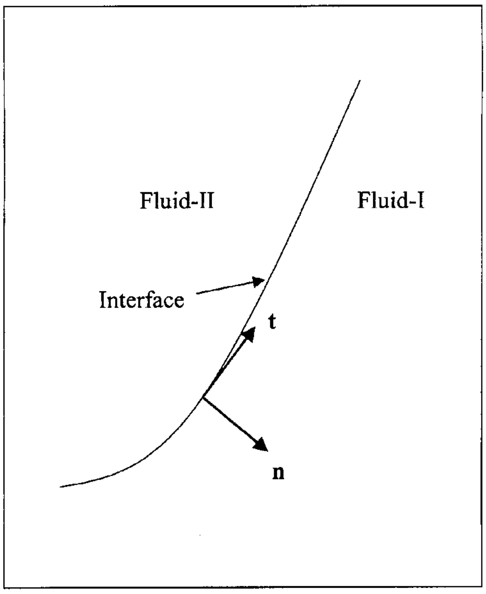

Fig. 1. Schematic diagram of fluid-fluid interface and coordinate system in the stratified flow.

the following conditions should be satisfied along the interface.

- kinematic condition: $\mathbf{v}\cdot\mathbf{n}=0$, $\mathbf{v}_I=\mathbf{v}_{II}$
- normal stress condition: $-p_I+(\tau_{nn})_I=-p_{II}+(\tau_{nn})_{II}$
- tangential stress condition: $(\tau_{nt})_I=(\tau_{nt})_{II}$

where n and t mean the normal and tangential directions at the interface, and the subscript I and II stand for the fluid-I and fluid-II, respectively. In the normal stress condition, surface tension effect is neglected, which is generally acceptable due to the high viscosity of polymer melts.

In the coextrusion problem the interface position is not known a priori. Thus, the interface position should be determined as part of solution. In this work the interface position is decoupled and treated separately from the flow field. Interface is updated using the previously calculated velocity profile (Nickell *et al.*, 1974). In the coextrusion, above set of equations is applied in each flow domain. Thus, the full set of equations for the coextrusion of two fluids is the combination of equations applied in each domain using the interface conditions.

The elastic viscous split stress (EVSS) method (Rajagopalan *et al.*, 1990) is used to deal with the viscoelastic stress terms. In EVSS method extra stress tensor is divided into the elastic and the viscous part, i.e. extra stress is rewritten as follows:

$$\boldsymbol{\tau}=\boldsymbol{S}+2\eta\boldsymbol{D} \tag{5}$$

where $\boldsymbol{S}$ is the modified extra stress tensor and $\boldsymbol{D}$ is the deformation rate tensor, which is expressed by

$$\boldsymbol{D}=(\nabla\boldsymbol{v}+\nabla\boldsymbol{v}^T)/2 \tag{6}$$

In using the EVSS method, the set of equations for the flow of UCM fluid therefore becomes:

$$\nabla\cdot\boldsymbol{v}=0 \tag{7}$$

$$\rho\boldsymbol{v}\cdot\nabla\boldsymbol{v}=-\nabla p+\nabla\cdot\boldsymbol{S}+\eta\nabla^2\boldsymbol{v} \tag{8}$$

$$\boldsymbol{S}+\lambda(\overset{\nabla}{\boldsymbol{S}}+2\eta\overset{\nabla}{\boldsymbol{D}})=0 \tag{9}$$

$$\boldsymbol{D}=(\nabla\boldsymbol{v}+\nabla\boldsymbol{v}^T)/2 \tag{10}$$

The Galerkin finite element method is used to discretize the above set of differential equations. The unknown velocity, pressure, modified stress, and deformation rate are approximated in terms of Galerkin basis functions:

$$\boldsymbol{v}=\sum_i\psi_i\boldsymbol{v}_i,\ p=\sum_i\phi_i p_i \tag{11}$$

$$\boldsymbol{S}=\sum_i\psi_i\boldsymbol{S}_i,\ \boldsymbol{D}=\sum_i\phi_i\boldsymbol{D}_i \tag{12}$$

where ϕ_i is the bilinear basis function and ψ_i is the biquadratic function. The governing equations, weighted integrally with the basis functions and integrated by parts, result in the weak form as follows:

$$\langle\boldsymbol{S}+\lambda(\overset{\nabla}{\boldsymbol{S}}+2\eta\overset{\nabla}{\boldsymbol{D}});\ \psi\rangle=0 \tag{13}$$

$$\rho\langle\boldsymbol{v}\cdot\nabla\boldsymbol{v};\psi\rangle+\langle-p\boldsymbol{I}+\boldsymbol{S}+\eta(\nabla\boldsymbol{v}+\nabla\boldsymbol{v}^T);\ \nabla\psi\rangle$$
$$=\langle\langle(-p\boldsymbol{I}+\boldsymbol{S}+\eta(\nabla\boldsymbol{v}+\nabla\boldsymbol{v}^T))\cdot\boldsymbol{n};\ \psi\rangle\rangle \tag{14}$$

$$\langle\nabla\cdot\boldsymbol{v};\ \phi\rangle=0 \tag{15}$$

$$\langle\boldsymbol{D}-(\nabla\boldsymbol{v}+\nabla\boldsymbol{v}^T)/2;\ \phi\rangle=0 \tag{16}$$

where $\boldsymbol{I}$ is the unit tensor and $\mathbf{n}$ is the outward normal vector. In the above equations, symbol < ; > and << ; >> represent domain and boundary line (or surface) integrals, respectively.

In the coextrusion process, the pressure and extra stresses are discontinuous along the interface. Therefore, special method should be used to treat this discontinuity. To treat the discontinuity of pressure and extra stresses across the interface, double-node finite element method has been used (Matsunaga *et al.*, 1998; Mavridis *et al.*, 1987). In the double-node method, the primitive variables such as extra stresses and pressure have two values at the same spatial location. The schematic diagram of grid used in double-node finite element method is shown in Fig. 2.

In the finite element method, the boundary integral part in Eq. (14) is generally replaced by the boundary con-

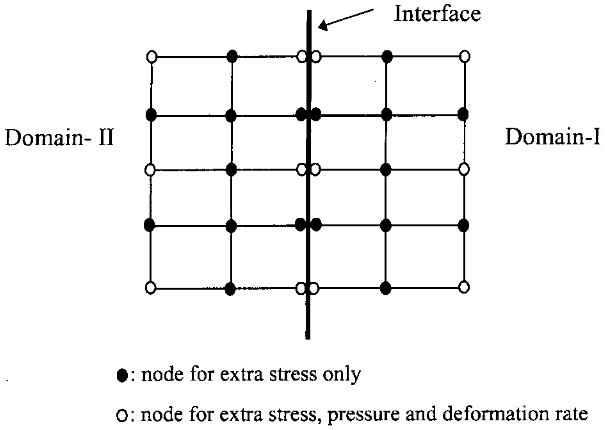

Fig. 2. Schematic diagram of the mesh used in the double-node finite element method.

ditions, either essential or natural. The boundary conditions should be compatible with the solution calculated within the domain of interest. However, if the accurate solution at the boundary is not known a priori, such as in the outflow boundary in the coextrusion of viscoelastic fluids or the nonisothermal viscoelastic flow, then it is almost impossible to impose a proper boundary condition. In the open boundary condition method, boundary integral parts are treated as part of unknown equations to be solved. Therefore the imposition of boundary condition along the outflow is not required. The components of the stiffness matrix related to the outflow boundary need to be modified to accommodate the boundary integral terms. Boundary nodal values are then obtained as part of the solution in this method.

To avoid the loss of convergence at high Weissenberg number, the streamline upwinding (SU) method is used to treat the convective terms in the constitutive equation (Marchal and Crochet, 1987). The set of nonlinear algebraic equations is then finally solved by means of Newtons method and the frontal elimination method.

3. Results and discussion

Schematic diagram of the coextrusion process is depicted in Fig. 3. Two fluids flow in each channel before the confluent point, after which two fluids merge and form the stratified flow. Initial finite element meshes used in this study are given in Fig. 4. Two meshes are identical except the downstream length. The boundary and interface conditions imposed in the simulation are as follows:

(i) at the inflow boundary (fluid-I and fluid-II)
 – fully developed velocity and extra stress profiles
(ii) at the die wall
 – no slip condition: **v=0**
(iii) at the outflow boundary

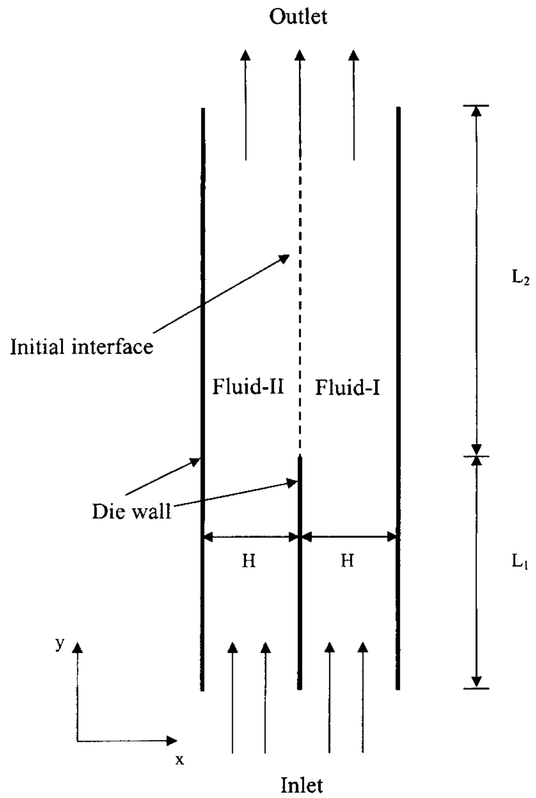

Fig. 3. Schematic diagram of the flow domain used in this study.

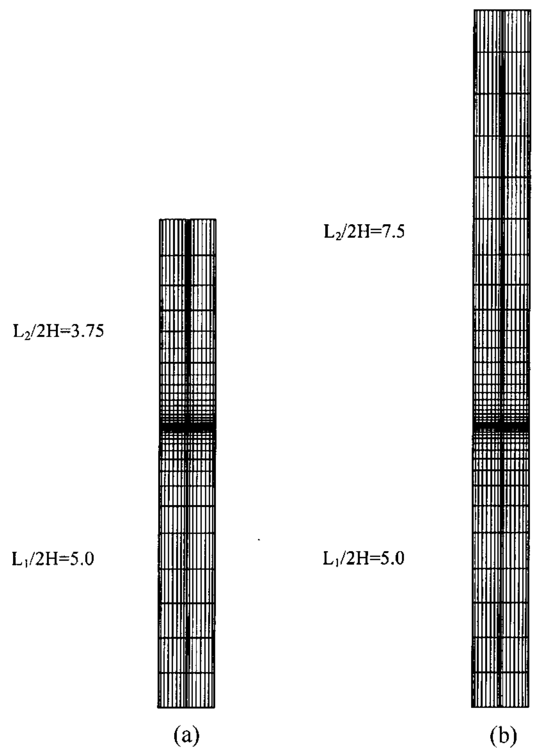

Fig. 4. Initial meshes used in this study: (a) MESH1 and (b) MESH2. Two meshes are identical except the downstream length.

– zero normal traction and tangential velocity
– zero normal and tangential tractions
– open boundary condition (OBC)

(iv) at the interface
– continuity of velocity
– continuity of total stress

Three kinds of boundary conditions are used at the outlet and the results obtained with each boundary condition are compared. Following dimensionless parameters are employed to describe the flow conditions of coextrusion of viscoelastic fluids.

Viscosity Ratio (VR) = η_{II}/η_I
Flow Rate Ratio (FRR) = Q_{II}/Q_I
Weissenberg number (We) = λ<v>/H

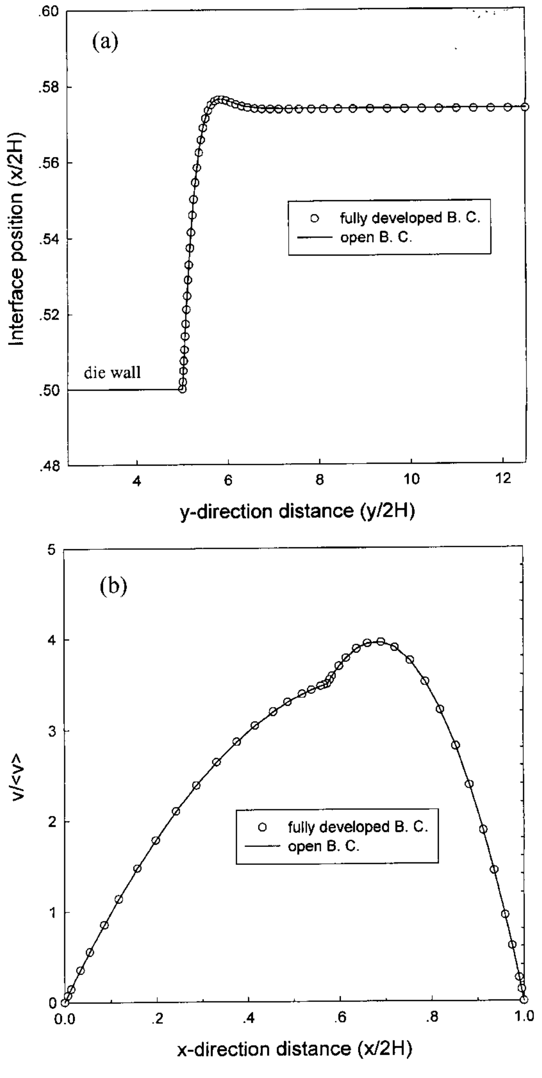

Fig. 5. Comparison of results obtained using the fully developed condition and those using the open boundary condition for Newtonian fluids at VR=5 and FRR=1 in MESH2: (a) interface position and (b) velocity profile at the outlet.

where η is the viscosity, Q is the flow rate, λ is the relaxation time, <v> is the average velocity of fluid in the separate channel, and H is the gap of channel before the confluent point.

3.1. Newtonian fluid case

In order to examine the reliability of open boundary condition method, we compared the results obtained using OBC with those using fully developed boundary condition, i.e. zero tangential velocity and zero normal traction, which has been conventionally used for generalized Newtonian fluids. In the case of generalized Newtonian fluids, the fully developed condition can be used without difficulty in the coextrusion simulation because the velocity profile need not be imposed essentially at the outflow boundary. Interface position and velocity profile calculated in MESH2 at VR=5 and FRR=1 are plotted in Fig. 5. As shown in the figure, velocity profile obtained using OBC is in good agreement with that obtained using the fully developed condition, and the interface position with the two boundary conditions is almost the same. These results show that the OBC method can be applied in the numerical simulation of coextrusion process.

To show the effect of viscosity ratio on the interface position and velocity profile at the outlet, numerical simulations are performed at various viscosity ratios and FRR=1 in MESH2. Deformed meshes at each viscosity ratio are shown in Fig. 6. The interface position and the outflow velocity variation with the viscosity ratio are also shown in Fig. 7. As the viscosity ratio increases, the interface position shifts toward fluid-I and the velocity of fluid-I increases

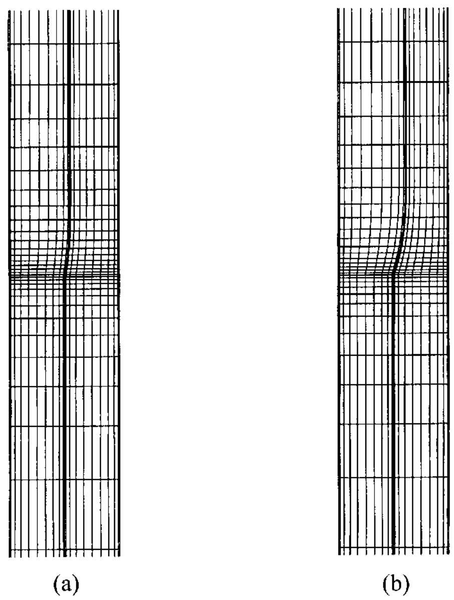

Fig. 6. Deformed meshes obtained at (a) VR=3 and (b) VR=10 Flow rate of two fluids is equal.

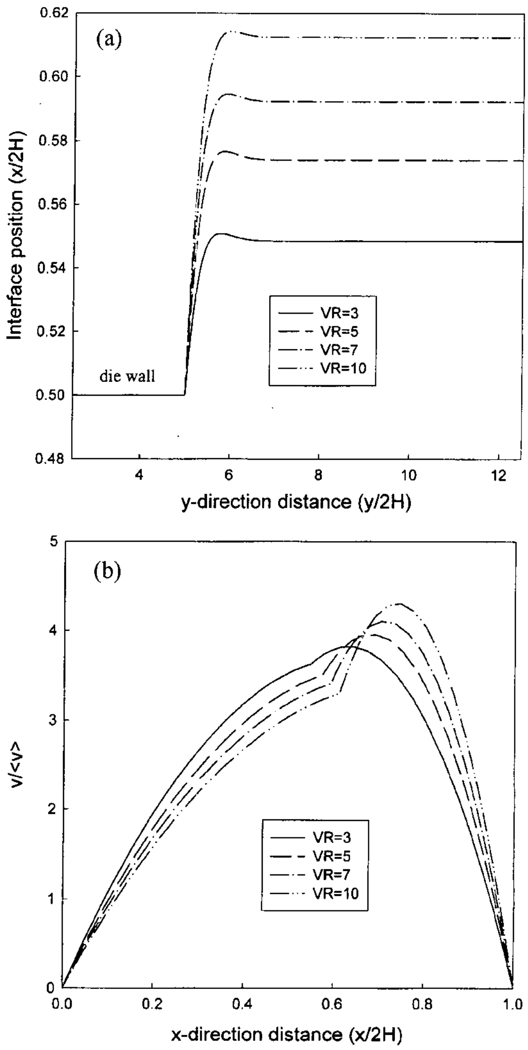

Fig. 7. Effect of viscosity ratio on (a) interface position and (b) velocity profile at the outlet in MESH2. Flow rate of two fluids is equal.

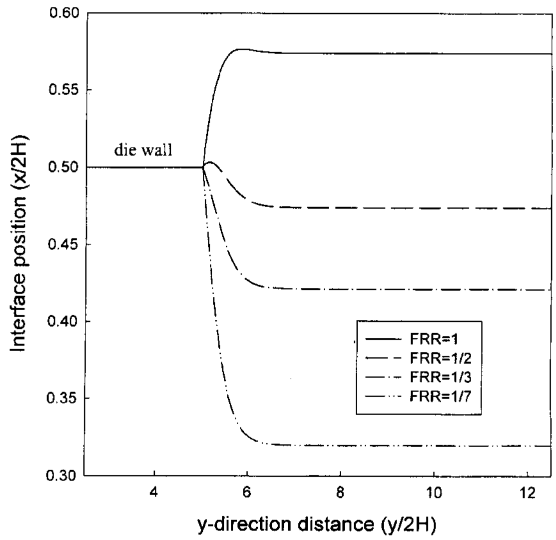

Fig. 8. Effect of flow rate ratio on the interface position in MESH2 (VR=5).

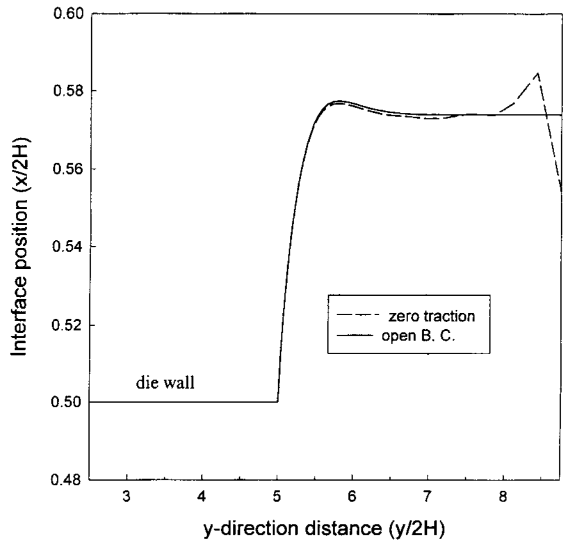

Fig. 9. Comparison of interface position calculated using the zero traction and the open boundary condition in MESH1 (VR=5, FRR=1 and We_I=0, We_{II}=0.25).

because fluid-I is easier to deform than fluid-II.

Interface positions at various flow rate ratios and VR=5 in MESH2 are plotted in Fig. 8. Fig. 8 shows that if the flow rate of two fluids is different, flow rate is more dominant than viscosity ratio in determining the interface position (Takase, 1998).

3.2. Viscoelastic fluid case

In order to show the outflow boundary condition effect on the interface position, we compare the results obtained using OBC method with those using zero normal and tangential traction condition, which were used by Matsunga *et al.* (1998). Interface positions obtained in MESH1 at VR=5, FRR=1, We_I=0 and We_{II}=0.25 are shown in Fig. 9. Here, We_I and We_{II} represent the Weissenberg numbers based on the flow conditions and the relaxation times of fluid I and II, respectively. The shape of interface position shows that the effect of outflow boundary condition is very important. As shown in Fig. 9, the interface position shows oscillatory shape at the outlet in the case of zero traction condition. Since zero traction condition is imposed at the outflow boundary, velocity and stress components should be changed to satisfy this boundary condition. Although the elasticity of fluid-II is not too high, the interface position is severely distorted near the outlet. In the case of OBC, on the contrary, the shape of interface position is smooth and does not change near the outlet. In OBC method, because outflow boundary condition is not imposed and velocity and stress components at the outflow boundary are calculated as part of solution, solutions calculated at outlet can be

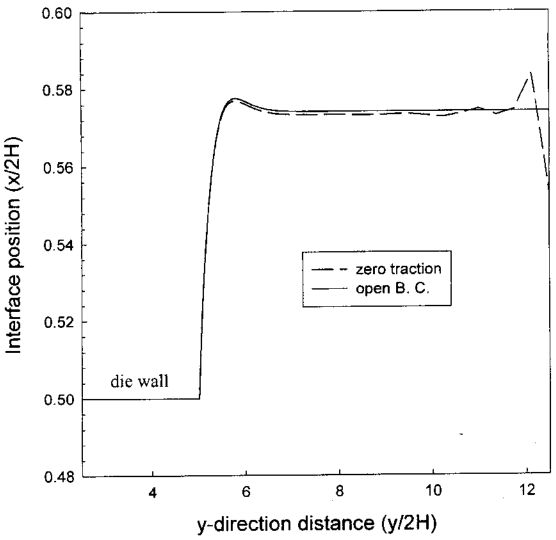

Fig. 10. Comparison of interface position calculated using the zero traction and the open boundary condition in MESH2 (VR=5, FRR=1 and We_I=0, We_{II}=0.25).

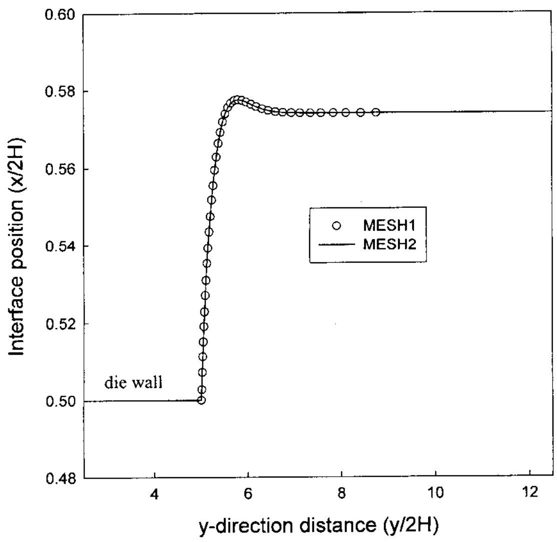

Fig. 11. Comparison of interface position calculated using the open boundary condition in MESH1 and MESH2 (VR=5, FRR=1 and We_I=0, We_{II}=0.25).

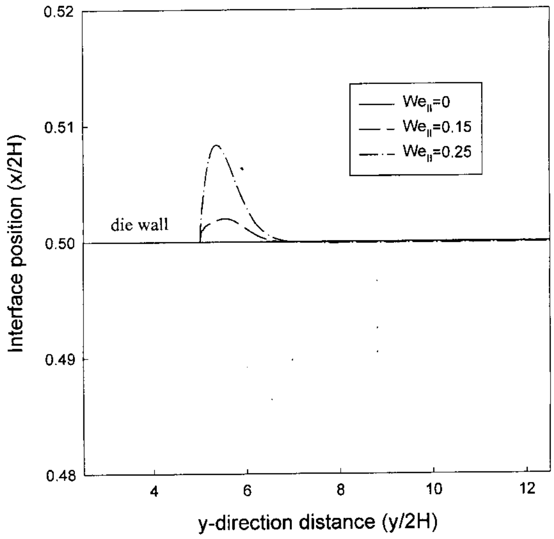

Fig. 12. Effect of elasticity of fluid-II on the interface position in MESH2 (VR=1, FRR=1 and We_I=0).

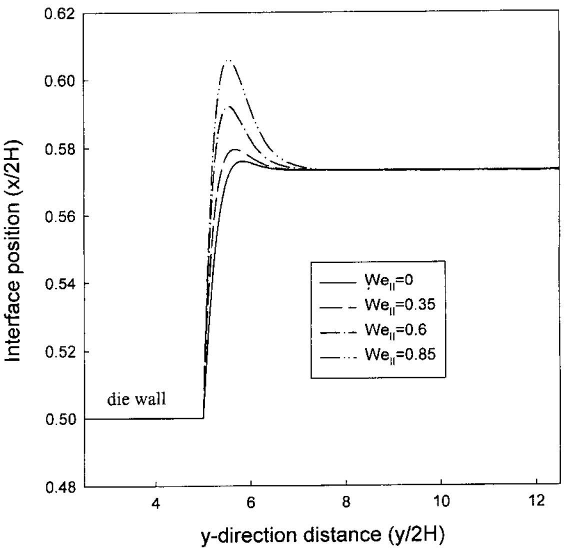

Fig. 13. Effect of elasticity of fluid-II on the interface position in MESH2 (VR=5, FRR=1 and We_I=0).

matched with those obtained at upstream.

To investigate the effect of downstream length on the interface shape, we carry out the simulation at the same condition in MESH2. The results are given in Fig. 10. In the case of zero traction condition, oscillatory shape of interface position also appears. However, if the results up to y/2H=8.5 are considered, two interface positions are not so different. That is, if long domain is used for the simulation using the zero traction condition, results corresponding to region away from the outlet are acceptable. In the case of OBC, as shown in Fig. 11, two interface shapes obtained in MESH1 and MESH2 are almost the same. This shows the reliability of the open boundary condition method in the simulation of coextrusion of viscoelastic fluids.

To understand the effect of elasticity on the interface position, numerical simulations are performed with changing the relaxation time of fluid-II at VR=1, FRR=1, and We_I=0 in MESH2. The interface positions at various Weissenberg numbers are shown in Fig. 12. From these results it is found that the first normal stress difference hardly affects the final interface shape. Experimental results also showed that the viscosity difference is more dominant than the first normal stress difference in the final interface shape (Han, 1981). However, the overshoot of interface position

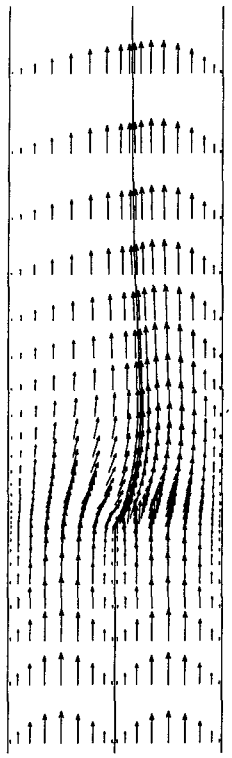

Fig. 14. Velocity vector plot at VR=5, FRR=1 and We_I=0, We_{II}=0.85 in MESH2.

appears near the confluent point and increases with elasticity. Therefore, if the length of die used in coextrusion is too short to reach the final interface position, the effect of elasticity is important in determining the interface position.

The interface positions at VR=5, FRR=1, and We_I=0 with increasing Weissenberg number of fluid-II are plotted in Fig. 13. Fig. 13 indicates that if the viscosity of two fluids is not equal, the interface shape near the confluent point is more dependent upon the elasticity of fluid due to the drastic velocity change near the confluent point. Velocity vectors at VR=5, FRR=1, We_I=0, and We_{II}=0.5 are shown in Fig. 14. Fig. 14 also shows the overshoot of interface and the drastic velocity change near the confluent point.

4. Conclusions

Numerical simulation of coextrusion process of viscoelastic fluids has been carried out inside a die. By employing the open boundary condition method, difficulty of imposing the boundary condition at the outflow boundary could be avoided. For the Newtonian fluid, the results obtained using the fully developed condition and the OBC showed similar profiles. In the case of UCM fluids, the results calculated with OBC showed the same interface shape in two meshes having different downstream length in contrast to those of zero traction condition. In addition, the interface position experiences oscillation close to the outlet in the case of zero traction. These results indicated that the open boundary condition method could be successfully applied in the numerical simulation of coextrusion process of viscoelastic fluids.

Viscosity difference was found to be more important than elasticity difference in determining the final interface position. However, the overshoot of the interface near the confluent point increased with the elasticity.

In this study, two-dimensional simulation was performed, in which side wall effect was not considered. Three-dimensional simulation will be necessary for understanding the encapsulation phenomena usually observed experimentally.

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