

# Coupled heat and mass transfer in laminar flow, tubular polymerizers

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**Abstract**—Thermal polymerization in a continuous flow, tubular reactor is largely determined by coupled heat and mass transfer processes. A detailed analysis is very important for the understanding of the transport phenomena involved and for the design and stable operation of such reactors. Computer experiments with this highly coupled, nonlinear system showed that for moderate flow rates in tubes larger than 4 cm in diameter, steep radial gradients in all principal variables and ultimately thermal runaway or flow channeling may occur. In this paper, the local interactions between strong temperature gradients, rapid polymerization, temperature/conversion-dependent transport properties and elongated velocity profiles have been analyzed for different thermal conditions.

## 1. INTRODUCTION

A PROTOTYPE of a continuous polymerization system would be essentially of a shell-and-tube type configuration. In such a process, the styrene monomer is pumped to a stainless-steel heat exchanger containing a bundle of tubes in a shell. The coolant flows through the shell side to maintain isothermal wall conditions. Alternatively, the tube walls are adiabatic. The monomer is heated to the desired inlet temperature in a preheater before entering the system. The threshold value for a thermally induced conversion of a monomer to a polymer is about 100°C. The rate and extent of reaction depends upon the local temperature and the residence time of the monomer. In turn, these two parameters are largely determined by the velocity field which is influenced by the fluid properties which in turn are temperature and concentration dependent. Assuming that the flow through all tubes of the prototype is identical, a single tube is representative of the entire system. Hence, the process consists of developing, laminar flow of a non-Newtonian fluid through a tube with exothermic reaction where an inlet monomer is (partially) polymerized. The fluid and thermal properties vary with temperature and monomer fraction.

Polymerizing mixtures are highly viscous with very low thermal conductivities and significant heat generation from the exothermic chemical reactions. If the generated heat is not removed rapidly enough, a 'hot spot' may occur leading to thermal ignition ( $T_{\text{crit}} \approx 245^\circ\text{C}$ ). Traditionally, the occurrence of a local hot spot along with the presence of a positive second derivative in the profile of temperature vs conversion before the hot spot is reached, is called a 'thermal runaway' condition for the reactor. For practical reasons, we defined 'thermal instability' as the occurrence of  $T \geq T_{\text{max}} = 200^\circ\text{C}$  anywhere in the reactor. Besides thermal instability, 'flow channeling' might occur. Considering a monomer molecule entering at, say, 130°C, a tube with isothermal walls of, say,

100°C, two opposing effects can be observed. In the tube center (core region) the temperature is higher whereas the time of contact is smaller when compared to temperature and exposure time near the wall (annular region). As a result, a large annular fraction of the tube may be occupied by a very slowly moving polymer whereas the core of the (monomer) solution would flow rapidly about the centerline. These flow patterns give rise to two average residence times which differ greatly if higher inlet temperatures and chemical initiators (for faster polymerization) or static mixers (for a more uniform distribution) are not being used. Furthermore, these flow conditions may lead to operational problems when the reactor can no longer be operated either because it has been clogged with polymer or because the monomer is getting through the tube largely unreacted due to channeling.

In a complementary paper [1] we determined the regions of stable reactor operations for a variety of boundary conditions, and we studied the parametric sensitivity on the conversion rate. It was found that a tube of radius up to 2 cm can be unconditionally used for continuous-flow polymerization. However, above this radius, thermal runaway, channeling and very large radial gradients in temperature and conversion may develop. Most previous theoretical investigations [2-4] assumed fully developed Poiseuille flow or a uniform velocity profile (i.e. plug flow), in order to concentrate more easily on stability aspects [2], reactor optimization [3] or multiple reaction kinetics [4]. In a less restrictive paper, Sala *et al.* [5] analyzed styrene polymerization in an adiabatic reactor assuming the fluid to be Newtonian. A first-order kinetics equation along with a constant molecular weight of 70,000 was employed.

In this paper, a comprehensive analysis is presented for coupled heat and mass transfer processes of steady laminar flow of a non-Newtonian fluid undergoing exothermic reaction in a straight tube with different thermal boundary conditions. Although this investigation is based on the process dynamics of styrene

## NOMENCLATURE

$c_p$	specific heat
$D_m$	mass diffusivity
$\Delta H$	heat of reaction
$k$	thermal conductivity
$L$	length of the tube
$M$	monomer concentration
$MW$	molecular weight
$p$	pressure
$Q$	flow rate
$r$	radial distance
$R$	radius of tube
$R_p$	polymerization rate
$R_w$	weight average chain length
$S_\phi$	source term related to principal variable $\phi$

$t$	time
$T$	temperature
$v_r$	radial velocity
$v_z$	axial velocity
$w_m$	monomer weight fraction
$w_p$	polymer weight fraction
$X_m$	conversion of monomer
$z$	axial distance.

## Greek symbols

$\rho$	density
$\dot{\gamma}$	rate of deformation
$\eta$	non-Newtonian viscosity
$\phi$	dependent variable
$\lambda$	diffusion coefficient.

polymerization, the model can be effectively used for a wide variety of basic studies in the engineering sciences.

## 2. MATHEMATICAL FORMULATION

## 2.1. The equation of motion

As mentioned earlier, the system consists of a straight circular tube which is maintained at isothermal or adiabatic wall conditions. The monomer solution is elevated to the desired inlet temperature in a preheater before entering the tube continuously with a parabolic velocity profile. The process is that of developing steady, laminar flow of a non-Newtonian fluid in a conduit with exothermic reaction where the inlet monomer is (partially) polymerized. The fluid properties vary with temperature and concentration.

Based on the stated assumptions, the equations of motion in cylindrical coordinates read (Fig. 1):

$$\text{(continuity)} \quad \frac{1}{r} \frac{\partial}{\partial r} (\rho r v_r) + \frac{\partial}{\partial z} (\rho v_z) = 0 \quad (1)$$

$$\begin{aligned} \text{(r-momentum)} \quad & \frac{1}{r} \frac{\partial}{\partial r} (\rho r v_r v_r) + \frac{\partial}{\partial z} (\rho v_z v_r) \\ & = -\frac{\partial p}{\partial r} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \eta \frac{\partial v_r}{\partial r} \right) + \eta \frac{v_r}{r^2} + \frac{\partial}{\partial z} \left( \eta \frac{\partial v_r}{\partial z} \right) \end{aligned} \quad (2)$$

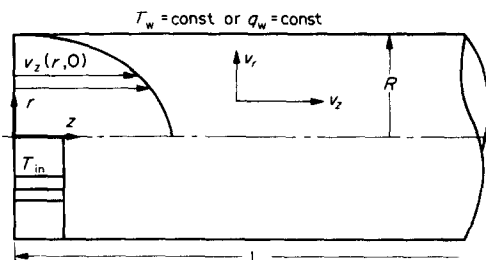


FIG. 1. System sketch.

$$\begin{aligned} \text{(z-momentum)} \quad & \frac{1}{r} \frac{\partial}{\partial r} (\rho r v_r v_z) + \frac{\partial}{\partial z} (\rho v_z v_z) \\ & = -\frac{\partial p}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \eta \frac{\partial v_z}{\partial r} \right) + \frac{\partial}{\partial z} \left( \eta \frac{\partial v_z}{\partial z} \right) \end{aligned} \quad (3)$$

$$\text{where } \eta = \begin{cases} \eta_0 & \text{for } 0 \leq \dot{\gamma} \leq \dot{\gamma}_0 \\ \eta_0 \left\{ 2 \left[ \left( \frac{\partial v_r}{\partial r} \right)^2 + \left( \frac{v_r}{r} \right)^2 + \left( \frac{\partial v_z}{\partial z} \right)^2 \right] \right. \\ \left. + \left( \frac{\partial v_r}{\partial z} + \frac{\partial v_z}{\partial r} \right)^2 \right\}^{(n-1)/2} & \text{for } \dot{\gamma} > \dot{\gamma}_0 \end{cases}$$

Here, a patched 'power law' has been employed as a suitable submodel for the polymeric solution [6]. In this power-law model, the low shear-rate viscosity,  $\eta_0$ , is equal to the viscosity correlation for the temperature- and concentration-dependent Newtonian fluid. The flow index,  $n$ , has an experimental value of 0.2 whereas  $\dot{\gamma}_0$  was found to be about  $5 \text{ s}^{-1}$  for polystyrene [7].

The boundary conditions include:

symmetry, i.e.  $v_r = 0$  and  $\partial v_r / \partial r = 0$  at  $r = 0$  for all  $z$ ;

no-slip, i.e.  $v_r = v_z = 0$  at  $r = R$  for all  $z$ ; entrance, i.e.  $v_z = 2v[1 - (r/R)^2]$ ; and

exit conditions, i.e.  $\partial v_r / \partial z = 0$  and  $\partial v_z / \partial z = 0$  for all  $r$ .

(4)

Density and viscosity are dependent upon temperature,  $T$ , polymer weight fraction,  $w_p$ , and average molecular weight,  $MW$  [1].

$$\rho = 1174.00 - 0.918T + (75.3 + 0.313T)w_p \quad (5)$$

$$\eta_0 = \exp \left[ -13.04 + 2103/T + (MW)^{0.18} (3.915w_p - 5.437w_p^2 + (0.623 + 1387/T)w_p^3) \right] \quad (6)$$

where

$$MW = 105R_w.$$

The local values of the scalar variables  $T$ ,  $w_p$ ,  $R_w$ , etc. are obtained from the solution of appropriate transport equations.

## 2.2. Transport equations

The general transport equation can be written for our axisymmetric system at steady state as

$$\frac{1}{r} \frac{\partial}{\partial r} (\rho r v_r \phi) + \frac{\partial}{\partial z} (\rho v_z \phi) = \frac{1}{r} \frac{\partial}{\partial r} \left( \lambda r \frac{\partial \phi}{\partial r} \right) + \frac{\partial}{\partial z} \left( \lambda \frac{\partial \phi}{\partial z} \right) + S_\phi \quad (7)$$

where  $\phi$  is the dependent variable,  $\lambda$  is the diffusion coefficient and  $S_\phi$  is a sink or source term. Each of the three parameters has to be defined for the auxiliary equations as shown below.

Equation of thermal energy:

$$\phi = c_p T, \quad \lambda = \frac{k}{c_p} \quad \text{and} \quad S_\phi = \Delta H R_p \quad (8)$$

where the heat of reaction  $\Delta H = 670 \text{ kJ kg}^{-1}$  and  $R_p$  is the polymerization rate [8]. The value for the specific heat was taken as  $c_p = 1880 \text{ J kg}^{-1} \text{ K}^{-1}$  whereas the thermal conductivities,  $k$ , for the monomer, polymer and the mixture are temperature dependent [8]. The associated boundary conditions are:

$$\begin{aligned} T(r=R) &= T_0 \quad \text{or} \quad \partial T / \partial r (r=R) = 0; \\ \partial T / \partial r (r=0) &= 0; \\ \partial T / \partial z (z=L) &= 0 \quad \text{and} \quad T(z=0) = T_i. \end{aligned} \quad (9)$$

Monomer mass transfer equation:

$$\phi = w_m, \quad \lambda = \rho D_m \quad \text{and} \quad S_\phi = R_p \quad (10)$$

where  $w_m$  is the monomer weight fraction,  $D_m$  is the mass diffusivity and  $R_p$  can be interpreted here as a sink term due to monomer consumption. The associated boundary conditions are:

$$\begin{aligned} \frac{\partial w_m}{\partial r} &= 0 \quad \text{at} \quad r=0 \quad \text{and} \quad r=R; \\ \frac{\partial w_m}{\partial z} &= 0 \quad \text{at} \quad z=L. \end{aligned} \quad (11)$$

Molecular weight equation:

$$\phi = w_p R_w, \quad \lambda = \rho D_m \quad \text{and} \quad S_\phi = -R_p R_w \quad (12)$$

where  $w_p$  is the polymer weight fraction and  $R_w$  is the weight average chain length [2] so that  $w_p R_w$  is the average weight of the polymer and  $S_\phi$  is here the source term for the molecular weight due to the newly formed polymer.

The associated boundary conditions are:

$$\begin{aligned} \frac{\partial}{\partial r} (w_p R_w) &= 0 \quad \text{at} \quad r=0 \quad \text{and} \quad r=R; \\ \frac{\partial}{\partial z} (w_p R_w) &= 0 \quad \text{at} \quad z=L. \end{aligned} \quad (13)$$

In summary, three transport equations for temperature, monomer weight fraction and average molecular weight of the polymer have to be solved with appropriate submodels representing reaction kinetics and changes in polymer chain length to obtain the local density and viscosity values [cf. equations (5) and (6)] in order to solve equations (1)–(3).

## 3. METHOD OF SOLUTION

The steady-state, two-dimensional momentum, heat and mass transfer equations subject to the associated boundary conditions have to be solved numerically. The solution technique employed here is the SIMPLE algorithm [9]. The calculation domain is divided into a number of non-overlapping control volumes, each finite volume surrounding one grid point. The differential equations are integrated over each control volume. Piecewise profiles expressing the variation of the dependent variables between grid points are used to evaluate the required integrals. The result is the discretization equation containing the values of  $\phi$ , etc. for a group of grid points. This algorithm uses an iterative procedure to solve the momentum equation for the staggered control volume mesh. While fluid flow conditions ( $p$ ,  $T$ ,  $w$ ) are evaluated at cell centers, the velocities are computed at cell boundaries. The mesh selected has 20 grid points in radial direction,  $0 \leq r \leq R$ , and 100 in axial direction,  $0 \leq z \leq L$ . With an initial guess for all unknowns ( $v$ ,  $p$ ,  $T$ ,  $w$ ) the program calculates the coefficients for each node of the discretized momentum equation. The velocity field, obtained from the momentum equation, will most likely not satisfy the continuity equation which in turn is then employed to develop a pressure correction equation. This pressure correction equation is solved and the velocity values updated to satisfy continuity. This velocity field is used to solve the discretized transport equations for the temperature, monomer and polymer distributions. Once the scalar quantities are determined, the fluid properties are updated. The values of all the unknowns are then compared to the previous values to check for convergence.

## 4. RESULTS AND DISCUSSION

### 4.1. Comparison with measured data

Only one experimental data set for the thermal polymerization of styrene in a tube was found in the open literature; Valsamis and Biesenberger [10] reported the following details about their laboratory experiment. The reactor consisted of a 0.46 cm I.D., stainless-steel, helically coiled tube of a total length of 14.6 m immersed in an isothermal bath of very high temperature, i.e.  $T_{\text{wall}} = 160^\circ\text{C}$ . Preheated monomer at  $T_{\text{in}} = 160^\circ\text{C}$  was delivered by means of a metering pump. The residence time was 5.15 min and an overall conversion of 15% was achieved.

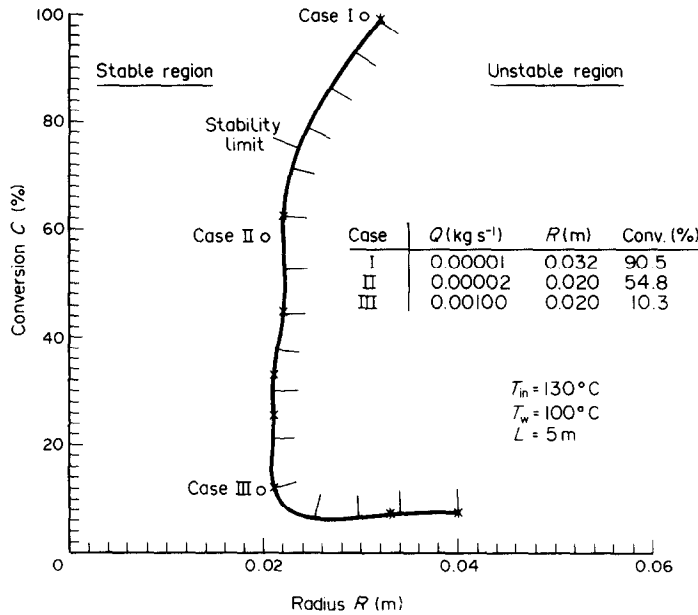


FIG. 2. Conversion-stability plot depicting three operational reactor cases.

Using the model developed in the present work, a test run with their system parameters in our *straight* tubular reactor yielded a conversion of 13.5%. This can be considered to be in good agreement with the experimental data considering the complex nature of the system being modeled. The kinetic rate expression, the viscosity and the other physical properties were taken from different sources (cf. [1]). Operation of this test reactor [10] was very close to the stability limit depicted in Fig. 2.

4.2. Temperature and monomer concentration profiles for three distinct operational cases

To analyze the coupled nonlinear momentum, heat and mass transfer in a tubular reactor characterized by

thermal polymerization processes, we selected three distinct cases, i.e. reactor operations at high, medium and low monomer conversions (cf. Fig. 2). The reactor of constant length and inlet temperature has isothermal walls of  $T = 100^\circ\text{C}$ . The conversion-stability plot of Fig. 2 is obtained by increasing the tube radius for a given mass flow rate until 'thermal runaway' occurs, i.e. the prescribed maximum temperature has been exceeded somewhere in the system. In running these computer experiments at different (constant) mass flow rates until thermal runaway occurs, different points for the stability limit can be obtained. Changes in inlet and/or boundary conditions shift or rotate the stability limit [2]. In Case I most of the incoming monomer is converted into polymer and such high conversion rates are only achievable with

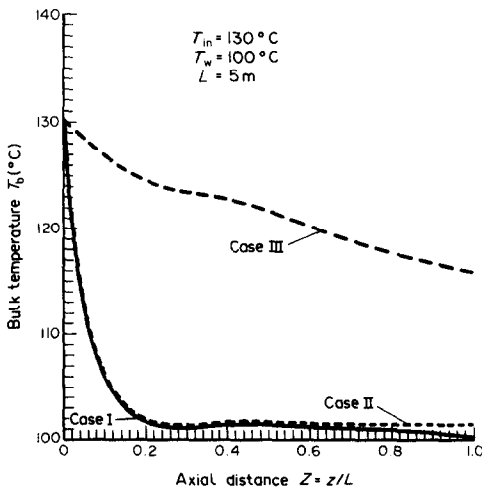


FIG. 3. Axial bulk temperature distribution for Cases I-III.

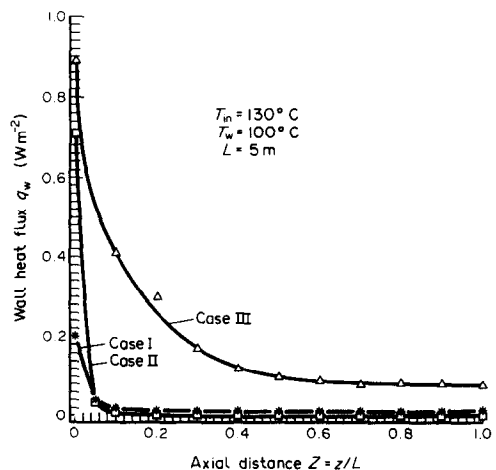


FIG. 4. Wall heat flux distribution for Cases I-III.

relatively small tubes and low flow rates. In contrast, Cases II and III feature flow rates which are substantially higher and monomer conversion rates which are average (Case II) or quite low (Case III). Figure 3 gives the bulk or 'mixing cup' temperature of the solution for the three cases. Cases I and II—characterized by low mass flow rates, i.e. very large exposure times—both show the rapid cooling of the polymeric liquid from the inlet temperature towards the wall temperature. In contrast, at higher mass flow rates (Case III), the almost unreacted monomer solution cools only gradually while moving through the reactor. The wall heat flux (Fig. 4), largely determined by  $T_{\text{wall}} - T_{\text{bulk}}$ , is almost zero for Cases I and II after a short 'entrance region'. The Nusselt number,  $Nu(z)$ ,

$$Nu = \frac{2q_w R}{k(T_w - T_b)} = -2R \frac{\partial T / \partial r|_w}{T_w - T_b}$$

is shown in Fig. 5 for the three cases. After rapid changes initially, an almost constant value of  $Nu \approx 2.5$  (cf. [7]) is maintained throughout the reactor length. In summary, variations in fluid parameters

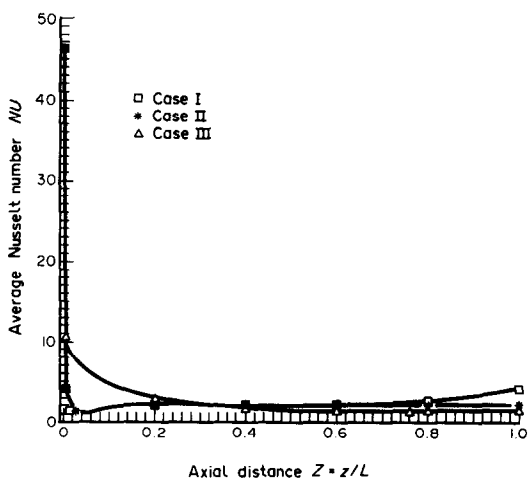


FIG. 5. Nusselt number distribution for Cases I-III.

and radial temperature gradients, as outlined in Section 1, account for the axial changes in integral properties  $T_b(z)$ ,  $q_w(z)$  and  $Nu(z)$ .

Figures 6a-c and 7a-c depict the radial temperature

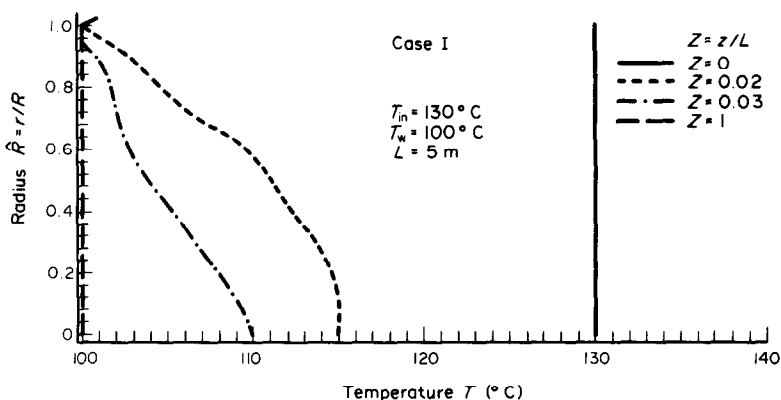


FIG. 6a. Radial temperature profiles at various stations for Case I.

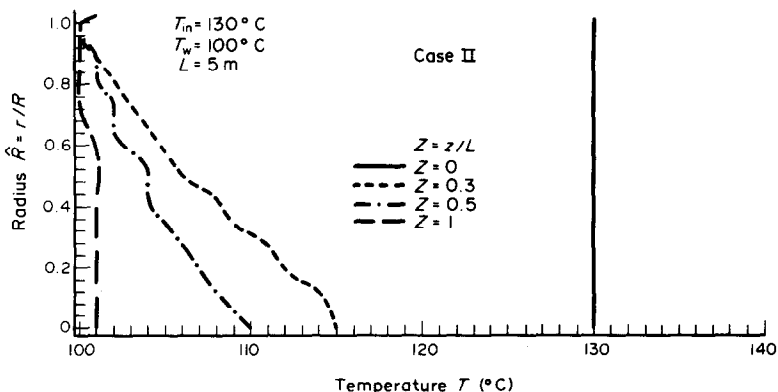


FIG. 6b. Radial temperature profiles at various stations for Case II.

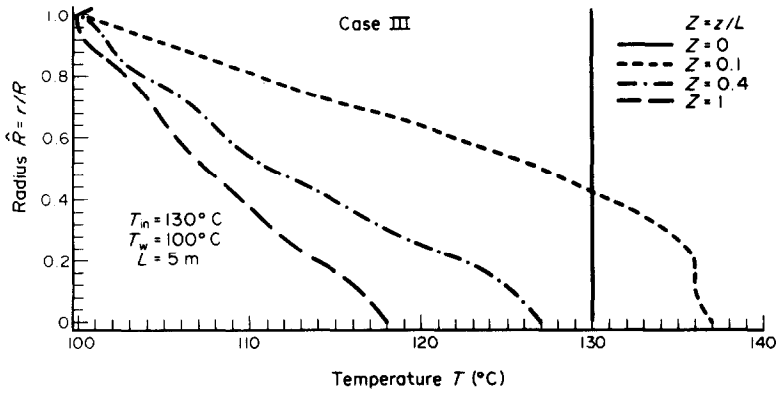


FIG. 6c. Radial temperature profiles at various stations for Case III.

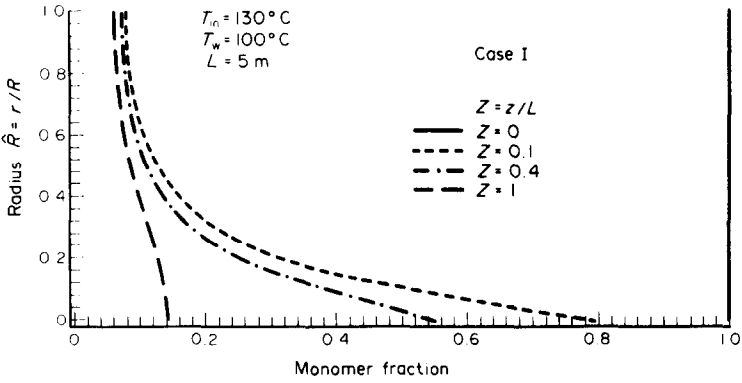


FIG. 7a. Monomer concentration profiles at various stations for Case I.

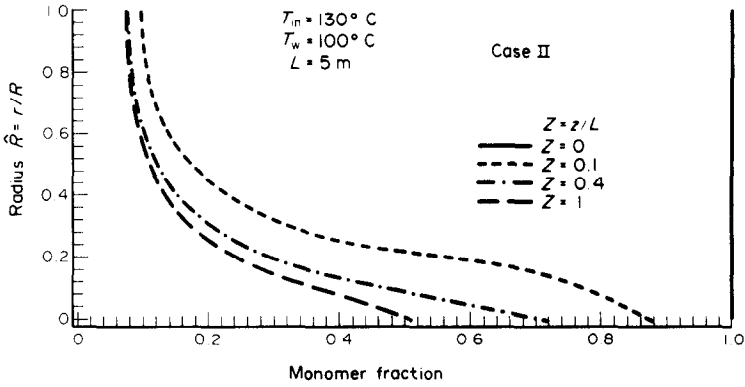


FIG. 7b. Monomer concentration profiles at various stations for Case II.

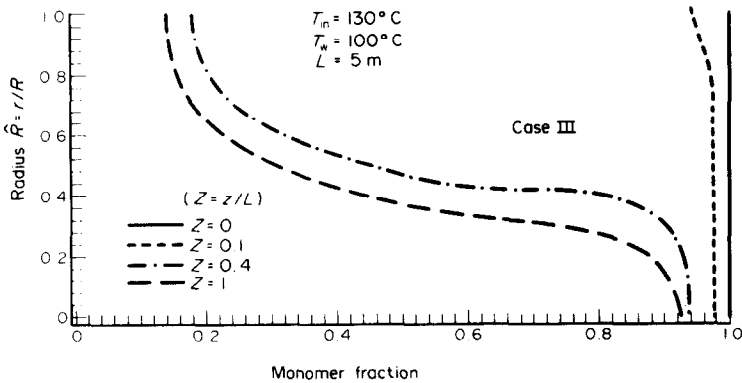


FIG. 7c. Monomer concentration profiles at various stations for Case III.

and monomer concentration profiles, respectively, for the three cases at various downstream stations  $Z = z/L$ . Because of the symmetry employed, only one half of the reactor is shown with  $\hat{R} = r/R = 0$  being the centerline and  $\hat{R} = r/R = 1$  being the tube wall. In accordance with Fig. 3, Cases I and II show a similar behavior (Figs. 6a and b) with a basically uniform exit temperature, especially in the case of high monomer conversion (Case I). Figure 6c indicates the high thermal inertia encountered with polymerization at low residence times. Near the tube entrance, when the monomer fraction is still high (cf. Fig. 7c), the core temperature exceeds the inlet temperature because of the exothermal reaction. Cooling mainly due to conduction takes place through the flow domain except for a thin wall layer formed near the tube exit (cf. Fig. 4). Figures 7a–c show the monomer concentration profiles,  $c(r, z)$ , and hence the two distinct regions, i.e. the partially unreacted monomer core and the annulus consisting mostly of polymer, as discussed in Section 1.

#### 4.3. Temperature and velocity profiles for adiabatic wall case

Somewhat comparable to Case III of Figs. 3 and 4, although opposite in trend, Fig. 8 shows the axial distribution of the wall temperature according to Newton's law of cooling. The radial temperature profiles for various axial stations in Fig. 9 demonstrate the effect of ideally insulated walls in conjunction with heat production due to chemical reaction. The two

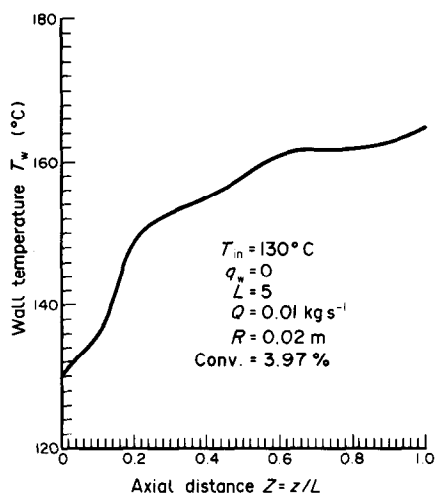


FIG. 8. Axial wall temperature distribution for adiabatic reactor.

regions of monomer core and polymer annulus appear again in Fig. 9 and even more pronounced in Fig. 10. Figure 10 shows the parabolic inlet profile as well as the development of the almost stagnant, i.e. highly viscous, polymer layer near the wall and the jetting effect of the greatly unreacted monomer about the centerline. Additional fluid dynamics aspects are presented in a separate paper [11].

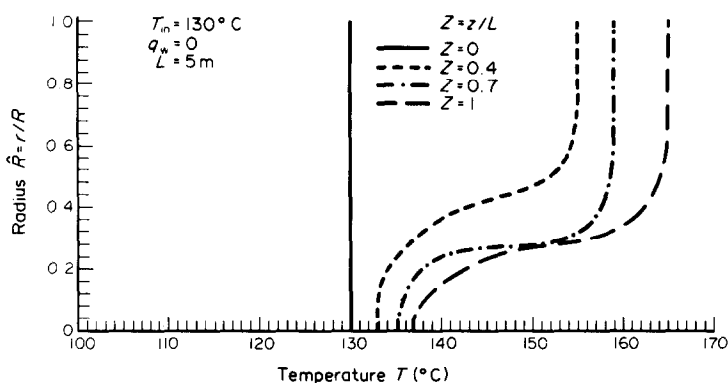


FIG. 9. Radial temperature profiles at various downstream stations for adiabatic reactor.

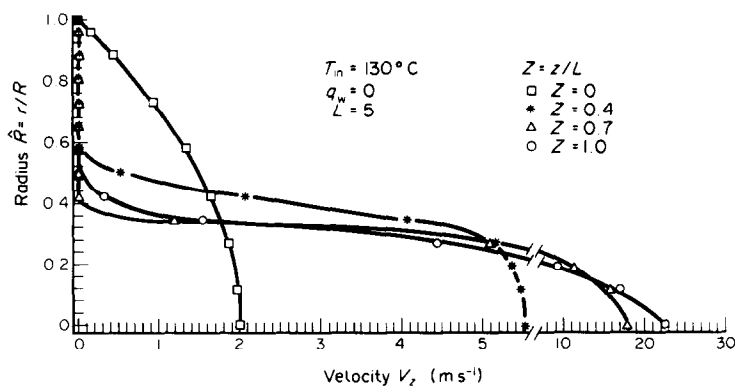


FIG. 10. Axial velocity profiles at various downstream stations for adiabatic reactor.

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TRANSFERT COUPLE DE CHALEUR ET DE MASSE EN ECOULEMENT LAMINAIRE  
DANS DES POLYMERISEURS TUBULAIRES

**Résumé**—La polymérisation thermique dans un réacteur continu est largement déterminée par les mécanismes couplés de transfert de chaleur et de masse. Une analyse fine est très importante pour la compréhension des phénomènes et pour la conception et le fonctionnement stable de tels réacteurs. Des essais numériques avec ce système non linéaire couplé montrent que pour des débits modérés dans des tubes de plus de 4 cm de diamètre, des gradients radiaux accentués peuvent apparaître pour toutes les variables principales. On analyse, pour différentes conditions thermiques, les interactions locales entre les forts gradients de température, la polymérisation rapide, les propriétés de transport variables et l'évolution des profils de température.

GEKOPPELTER WÄRME- UND STOFFÜBERGANG BEI LAMINARER STRÖMUNG  
IN ROHRFÖRMIGEN POLYMERISATIONS-REAKTOREN

**Zusammenfassung**—Die kontinuierliche thermische Polymerisation in rohrförmigen Reaktoren wird hauptsächlich durch die Vorgänge beim Wärme- und Stoffübergang bestimmt. Eine ausführliche Analyse ist für das Verständnis der Transportvorgänge, für die Auslegung und für den stabilen Betrieb solcher Reaktoren sehr wichtig. Rechner-Untersuchungen für dieses stark gekoppelte, nichtlineare System zeigten, daß in Rohren mit einem Durchmesser größer als 4 cm bei mäßigem Durchsatz sehr hohe Gradienten aller maßgeblichen Einflußgrößen in radialer Richtung auftreten, und dadurch letztendlich ein thermisches Durchgehen oder eine Kanalbildung auftreten kann. In dieser Abhandlung wird die örtliche Wechselwirkung zwischen hohen Temperaturgradienten, schneller Polymerisation, temperatur/umwandlungsabhängiger Transport-Eigenschaften und langgestreckten Geschwindigkeitsprofilen für verschiedene thermische Bedingungen untersucht.

ТЕПЛО-И МАССОПЕРЕНОС ПРИ ЛАМИНАРНОМ ТЕЧЕНИИ В ТРУБЧАТЫХ  
ПОЛИМЕРИЗАТОРАХ

**Аннотация**—Полимеризация в трубчатом реакторе непрерывного действия в значительной мере определяется взаимосвязанным тепло-и массопереносом. Для понимания механизмов переноса, а также конструирования и обеспечения надежной работы реакторов подобного типа необходим детальный анализ протекающих процессов. Численные эксперименты с нелинейной взаимосвязанной системой показали, что для средних скоростей течения в трубах с диаметром больше 4 см могут иметь место резкие радиальные градиенты всех основных переменных и конечных значений теплоотдачи или характеристик течения. Местные взаимодействия между резкими перепадами температуры, быстрая полимеризация, характеристики переноса, зависящие от температуры и глубины превращения, вытянутые профили скорости анализируются для различных тепловых условий.