MODEL OF THE CLOGGING OF CHANNELS WHEN POLYMERIZING LIQUIDS FLOW THROUGH THEM

924

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An analytical investigation is conducted into the flow of a viscous polymerizing liquid. The theoretical basis is given for the possibility of completely clogging sections of the channel after some time under conditions of constant flow of the reaction medium. The effect of transverse diffusion on the clogging process is considered. It is shown that, in spite of the small value of the molecular diffusion coefficient, transverse diffusion can markedly change the nature of the clogging process. The effect of transverse diffusion explains the cessation of the clogging under certain relationships of the diameter, the length, the flow rate, and the polymerization rate. A mathematical model of the process is presented, and relationships are obtained for evaluating the length of the input section and the formation rate of an stagnant layer at the walls.

The clogging of tubes when polymerizing liquids flow through them is encountered in many industrial processes: in tubular high-pressure polyethylene polymerizing machines, the circulating loops of polymerizing machines, and heat-transfer tubes and piping for making polystyrene plastics. If this effect is not considered, evaluations the effect of basic kinetic and hydrodynamic characteristics on the clogging rate and the design of similar processes are made difficult; thus the interest in this problem [1, 2]. From a qualitative viewpoint, explaining the clogging effect is not complicated. The degree of conversion of the monomer depends on the residence time in the flow, which tends to infinity as the flow approaches the wall. In turn, the viscosity depends strongly on the conversion; consequently a layer of enhanced viscosity (in the infinite limit) forms at the wall and gradually propagates to the center of the flow.

A relationship for the growth rate of this layer for a constant pressure head and for a constant material flow has been found [1, 2] in the approximation when the stagnant layer of reaction products is thin compared to the width of the channel. It is shown that for a constant pressure head the channel clogs up after a finite time, but for constant flow the radius drops off as 1/t, which does not agree with experiment, however. Thus, in making polystyrene plastics, total clogging of certain circulating and transport piping is observed in a finite time, when the liquid is fed by gear-type pumps, which create a mass flow that depends weakly on the pipe resistance.

The viscosity of the polymerizing liquid depends not only on the polymer concentration (conversion) and the temperature (the temperature dependence is substantial, because the polymerization reaction is exothermic), but also on the molecular-weight distribution. Therefore a total description of the pressure flow of such a liquid should include not only the dynamic equation for a non-Newtonian fluid with variable properties and a corresponding heat transfer equation, but also a detailed kinetic model for calculating the molecular characteristics of the forming polymer. Therefore the solution of this problem in its full formulation is extremely complex. However, a feature of most polymerization processes is the strong dependence of the viscosity of the reaction medium on time, because 1) the viscosity increases strongly with increasing conversion and 2) the reaction rate, as a rule, increases sharply with increasing viscosity (the jellium effect) [3]. For sufficiently slow reactions, where the flow can be considered isothermal, this greatly simplifies the analysis and makes it possible to obtain relationships for evaluating the growth of the stagnant polymer layer and the clogging time of the channel.

We assume [2] that the state of an element of the medium is determined by its residence time in the reaction volume, such that for $\tau \leq T$ it is a liquid and for $\tau > T$ it is a solid (T is the characteristic time for the polymerization reaction). In this formulation, the

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residence time τ and the shape of the boundary of the stagnant layer are found from the equation

$$d\tau/dt = 1 = \partial \tau/\partial t + \mathbf{V} \nabla \tau \tag{1}$$

(\mathbf{V} is the vector flow velocity and t is the time) with the boundary conditions $\tau = 0$ at the inlet to the reaction volume and $\tau = T$ at the boundary of the clogging stagnant layer.

We will solve this problem with the following assumptions: 1) the clogging rate is small and is determined by the velocity field only directly next to the solid boundary, which makes it possible to consider the velocity profile to be linear; 2) the thickness of the stagnant layer is much less than the characteristic dimension of the flow surface.

Initially we will examine the special case of a constant shear rate $\dot{\gamma}$ = const, which corresponds to longitudinal flow over a flat plate or flow in a channel of constant width (radius) at the initial stage of the process, while H \ll R (H is the thickness of the clogging layer and R is the channel radius). In this case, the system (1) takes the form

$$\partial \tau / \partial t + u \partial \tau / \partial x + v \partial \tau / \partial y = 1; \tag{2}$$

$$\tau|_{x=0} = 0, \ \tau|_{y=H(x,t)} = T, \tag{3}$$

where x is the distance from the beginning of the solid boundary; Y is the distance from the solid boundary along the normal, and u and v are the velocity components along and normal to the solid boundary.

From the linear approximation of the velocity profile, it follows that

$$u = \gamma(y - H(x, t)), \tag{4}$$

and from the continuity equation $\nabla V = 0$

$$v = \gamma (y - H(x, t)) \partial H / \partial x.$$
(5)

The solution to Eq. (2) in the region with a moving boundary is difficult; therefore we make a change of variables

$$\Theta = t - T, \quad Y = y - H(x, t), \quad X = x. \tag{6}$$

We substitute Eqs. (4)-(6) into (2) and (3) and obtain

$$\partial \tau / \partial \Theta + \gamma Y \partial \tau / \partial X - \varphi \partial \tau / \partial Y = 1, \ \tau|_{Y=0} = T, \ \tau|_{X=0} = 0$$
⁽⁷⁾

where $\phi(X, \Theta) = \partial H(X, \Theta)/\partial \Theta$ is the growth rate of the stagnant layer of reaction products. Equation (7) for the unknowns τ and ϕ is an equation in the stagnant region with constant coefficients and stationary boundary conditions. It is natural to assume that its solution approximately approaches the solution of the stationary problem

$$\gamma Y \partial \tau / \partial X - \varphi \partial \tau / \partial Y = 1; \tag{8}$$

$$\tau|_{X=0} = 0, \ \tau|_{Y=0} = T.$$
(9)

We show that this formulation is correct and makes it possible to find both $\tau(X, Y)$ and $\phi(X)$. We introduce the variable s(X, Y), which satisfies the conditions

$$\partial Y/\partial s = \varphi(X), \ \partial X/\partial s = -\gamma Y;$$
 (10)

$$s|_{Y=0} = 0 \text{ or } Y|_{s=0} = 0.$$
 (11)

From (10), Eq. (8) takes the form $d\tau/ds = -1$ or

$$\tau = \tau|_{s=0} - s = \tau|_{Y=0} - s = T - s.$$
(12)

It follows from (9) and (12) that $\tau |_{X=0} - 0 = T - s$, that is,

$$s|_{X=0} = T.$$
 (13)

It is obvious that Eqs. (10) and (11), which are supplemented by the condition

$$X|_{s=0} = p \tag{14}$$

(p is an arbitrary positive number) for a given $\phi(X) > 0$, has a unique solution in the interval $X \in (0, p)$. The problem consists of finding the function $\phi(X)$ which fulfills the condition [13] for any p.

After dividing the first equation of the system (10) by the second, we obtain dY/dX =

 $-\phi(X)/(\dot{\gamma}Y)$. From this expression and (11) and (14) we obtain $\dot{\gamma}Y^2/2 = \int \phi(z) dz$ or

$$Y = \sqrt{\frac{2}{\dot{\gamma}} \int_{X}^{p} \varphi(z) dz}.$$
(15)

Substituting (15) into (10) yields $dX/ds = -\dot{\gamma} \sqrt{\frac{2}{\dot{\gamma}} \int_{X}^{p} \varphi(z) dz}$, from which

$$\int_{0}^{p} \frac{dX}{\sqrt[y]{\frac{2}{\gamma} \int_{X}^{p} \varphi(z) dz}} = \int_{\begin{pmatrix}X=p\\Y=0\end{pmatrix}}^{p} ds,$$
(16)

where p is a positive number.

From physical considerations, it is clear that $\phi \rightarrow 0$ for $z \rightarrow 0$. This makes it possible to seek a solution to the integral equation (16) in the form

$$\varphi(z) = az^b + O(z^b) \tag{17}$$

where O(X) is infinitely small compared to the magnitude of X. Substituting (17) into (16) and introducing the notation $\chi = X/p$ and $\eta = z/p$, we find

$$\int_{0}^{1} \frac{p \, d\chi}{\sqrt{2a\dot{\gamma} \, p^{b+1} \int_{\chi}^{1} \eta^{b} \, d\eta \left(1 + O\left(\frac{b+1}{p}\right)\right)}} = T.$$
(18)

The left side of Eq. (18) is independent of T only for b = 1, $O(p^{(b+1)/2})$. In this case

$$\int_{0}^{\cdot} \frac{d\chi}{\sqrt{a\dot{\gamma}(1+\chi^2)}} = \frac{\pi}{2\sqrt{a\dot{\gamma}}} = T, \quad a = \pi^2/(4T^2\dot{\gamma}), \quad \varphi(X) = \pi^2 X/(4T^2\dot{\gamma}).$$

Thus, the boundary separating the solution and the stagnant layer of reaction products rotates around the beginning of the plate with an angular velocity

$$\Omega = \pi^2/(4\gamma T). \tag{19}$$

We now examine the variable shear rate $\dot{\gamma} = \dot{\gamma}(X, \Theta)$. Here integration of the continuity equation for v gives

$$v = \dot{\gamma} (y - H) \frac{\partial H}{\partial x} - \frac{1}{2} \frac{\partial \dot{\gamma}}{\partial x} (y - H)^2 = \dot{\gamma} Y \frac{\partial H}{\partial x} - \frac{Y^2}{2} \frac{\partial \dot{\gamma}}{\partial X},$$
 (20)

instead of [5]. By substituting (20) into (1) we obtain

$$\dot{\gamma}Y\partial\tau/\partial X - \varphi\partial\tau/\partial Y = 1 - Y^2 \partial\gamma\partial\tau/(2\partial X\partial Y).$$
(21)

We now show that the solution to Eq. (8) for $\phi(x)$ is a lower estimate for the solution to Eq. [22]. Actually, now, at each point of the flow and at each moment in time, let the "aging" rate of the medium increase by a factor of $1 + \Psi(X, Y, \Theta)$ ($\Psi \ge 0$), that is, $d\tau/d\Theta =$ $1 + \Psi(X, Y, \Theta)$. This effect leads to the appearance of an additional term in Eq. (8):

$$\frac{1}{\gamma}Y\partial\tau/\partial X - \varphi\partial\tau/\partial Y = 1 + \Psi.$$
 (22)

At the same time, acceleration of the "aging" of the medium can lead only to an increased clogging rate. If we compare (21) with (22), we see that the term $-Y^2\partial\dot{\gamma}\partial\tau/(2\partial X\partial Y)(\partial\dot{\gamma}/\partial X > 0$ and $\partial\tau/\partial Y < 0$) now plays the role of Ψ .

For $Y \rightarrow \infty$, this quantity is of the order $\Delta \dot{\gamma}/(2\dot{\gamma})$. Simple evaluations also show that it decreases with decreasing Y and tends to zero near the stagnant layer of reaction products. That is, the solution to (21) can be estimated with sufficient accuracy by the solution to Eq. (8) (with a consideration of the dependence of $\dot{\gamma}$ on X).

If we substitute $d\tilde{X} = dX/\dot{\gamma}(X)$, Eq. (22) is reduced to the form $Y\partial\tau/\partial\tilde{X} - \phi\partial\tau/\partial Y = 1$,

and the solution $\phi = \pi^2 \tilde{X}/(4T^2)$, and consequently the local angular velocity of an element of the surface H(x, t), is first of all determined by the relationship

$$\Omega(X) = d\phi/dX = d\phi/(\gamma(X)dX) = \pi^2/(4\gamma(X)T^2).$$

If we express $\dot{\gamma}(x)$ in terms of the volume flow rate of the medium Q and the running radius of the tube r, we write the equation for the clogging of the tube:

$$\partial^2 r / (\partial X \partial t) = -\Omega = -\pi^3 r^3 / (16QT^2).$$
⁽²³⁾

If we go to the dimensionless variables $\rho = r/R$, $\tilde{l} = X/R$, $\tilde{t} = \pi^3 R^3 t/(16QT^2)$ (R is the radius of the tube), we obtain

$$\partial^2 \rho / \partial \tilde{l} \partial \tilde{t} = -\rho^3, \quad \rho \mid_{\tilde{l}=0} = 1, \quad \rho \mid_{\tilde{l}=0} = 1.$$
(24)

Because a change of scale in the quantities X and T, while conserving the derivative of these scales, does not change the form of Eq. (24), the solution to Eq. (24) is the function of a single variable $\eta = \tilde{\ell}t$. When η is introduced into Eq. (24), it takes the form

$$\eta d^2 \rho / d\eta^2 + d\rho / d\eta = d(\eta d\rho / d\eta) / d\eta = -\rho^3;$$
⁽²⁵⁾

$$\rho|_{\eta=0} = 0.$$
 (26)

The second initial condition is obtained from the requirement that the solution $\rho(\eta)$ to (25) be smooth and finite:

$$d\rho/d\eta|_{\eta=0} = -1. \tag{27}$$

The function $\rho(\eta)$, which is found from the numerical solution of Eq. (25) with the initial conditions (26) and (27), is shown in Fig. 1.

If we return to dimensional variables, we have the dependence of the tube radius as a function of time and coordinates: $r = \pi^3 R^3 \rho xt/(16QT^2)$, and also an expression for the pressure drop Δp in a tube of length $\tilde{\ell} = \eta/\tilde{t}$:

$$\Delta p / \Delta p_0 = \frac{1}{\eta} \int_0^{\eta} dx / \rho(x), \qquad (28)$$

which is shown in Fig. 1.

As opposed to [2], a feature of the resultant solution is that the function $\rho(\eta)$ does not tend asymptotically to zero as $\eta \rightarrow \infty$, but becomes zero at $\eta \approx 3$. The function in [2] is the result of assuming that the thickness of the stagnant layer of reaction products is small compared to the tube diameter. We only assumed that the partial derivative $\partial H/\partial x$ is small.

Once we take the dimensionless clogging time as $\eta = 3$ and return to a dimensional form, we obtain the expression for the clogging time:

$$t^* = 48QT^2/(\pi^3 R^2 l).$$
⁽²⁹⁾

The resultant expression correctly reflects the basic features of the process of clogging the channels. We note that Eqs. (19), (20), and (23), which relate the clogging rate with characteristic dimensions and the shear rate at the wall, can be applied without requiring a detailed description of the pressure flow throughout the whole reaction volume. Therefore they can be used to analyze more complex situations than in a channel, for example in analyzing the process of clogging the walls of a polymerizing machine which has a mixer. In this case, the characteristic shear rate can be determined from the relationship [4]

$$\gamma = M/\mu\pi Dh = K_N(\text{Re})\sigma n^2 d^5/2\pi^2 \mu Dh,$$

where σ and μ the density and viscosity of the medium; D and h are the diameter and height of the apparatus; d is the diameter of the mixer; n is the rotation rate of the mixer; Re = $nd^2\sigma/\mu$ is the Reynolds number; K_N is the power coefficient of the mixer; and M is the torque on the shaft of the mixer.

Either the characteristic dimension of the large-scale pulsations [5] or (for very slow flows) the length of the corresponding vortex streamline L ~ $hU/W \sim \pi hdD^2n/16Q$ plays the role of the section length X (Q is the circulation capacity of the mixer, and U and W are the average circular and axial velocities in the apparatus).





Analysis of (29) shows that proportionality of the clogging time t* to Q, T^2 and l^{-1} is natural. However, the proportionality of t* to the quantity R^{-2} is unexpected at first glance. The clogging time is less for a tube of larger radius than a smaller one. This would be impossible if the tube clogged uniformly along its length. However, the clogging rate of the channel in each section is governed by the flow history of the liquid in the preceding sections of larger radius. This result agrees qualitatively with the practical operation of piping in many polymer factories, where decreasing the piping diameter leads to a significant reduction of the [polymerization] rate, and - in a series of cases - to a total elimination of clogging of the piping surface.

Usually the effect of diffusion is neglected in analyzing laminar flow of the reacting media. As a rule, this approximation is correct because of molecular diffusion coefficients in liquids are small and, consequently, so is diffusion flow compared to convective flow. However, diffusion effects become substantial for the flow of a polymerizing liquid near the solid wall at the input section of the channel. The operational experience of a number of polymer factories shows that in some cases clogging does not continue indefinitely if the piping sections are not too long. We will show that this effect can be explained by transverse diffusion.

We augment Eq. (1) by a term which considers diffusion in the y direction:

$$yy\partial\tau/\partial x - \varphi\partial\tau/\partial y - D\partial^2\tau/\partial y^2 = 1,$$
(30)

where D is the molecular diffusion coefficient.

From physical considerations it is clear that the growth rate ϕ of an attached layer is zero sufficiently close to the input section (x = 0) (the diffusion thickness of the boundary layer tends to zero for x \rightarrow 0). In this section, Eq. (30) has the form

$$\gamma y \partial \tau / \partial x - D \partial^2 \tau / \partial y^2 = 1, \tag{31}$$

and the natural boundary conditions for this equation are

$$\tau|_{x=0} = 0, \quad \tau|_{y\to\infty} \to 0, \quad \frac{\partial \tau}{\partial y}\Big|_{y=0} = 0.$$
(32)

We take Eqs. (31) and (32) to infinity, introduce the variables $\theta = \tau/T_0$, Y = y/L, X = x/L, where $T_0 = \dot{y}^{-1}$, $L = \sqrt{DT_0}$ and obtain:

$$\partial^2 \Theta / \partial Y^2 - Y \partial \Theta / \partial X = -1, \ \Theta|_{X=0} = 0,$$

$$\Theta|_{Y \to \infty} \to 0, \ \partial \Theta / \partial Y|_{Y=0} = 0.$$
 (33)

After we take the Laplace transform of (33) with respect to X, it is easy to find the solution to the resultant ordinary differential equation. Then we take the asymptotic expansion of the resultant solution at the point Y = 0, take the inverse Laplace transform, and have

$$\Theta|_{Y=0} = \Gamma\left(\frac{1}{3}\right) / \left[3^{2/3}\Gamma\left(\frac{5}{3}\right)\right] \cong 1.4X^{2/3}.$$

We return to dimensional variables for the residence time at the solid surface and obtain $\tau(x) = (1,4/\gamma)(D/\gamma)^{-1/3}x^{2/3}$. Obviously, the coordinate x* of the point at which the clogging of the solid surface starts is determined by the condition $\tau = T$:

$$r^* = 0.58 \dot{\gamma} D^{1/2} T^{3/2} = (0.34DT)^{1/2} \dot{\gamma} T.$$
(34)

We estimate the length of the unclogged section x^* . We set $\dot{\gamma} = 10 \text{ sec}^{-1}$, $D = 10^{-10} \text{ m}^2/\text{sec}$, and $T = 10^3 - 10^4$ (e.g., the reaction time characteristic for conditions for producing polystyrene plastic). Then according to (34), $x^* = 2-60 \text{ m}$. Thus, the effect of transverse diffusion, in spite of the small value of D, can guarantee a significant input section of tubes and channels whose walls are not clogged by polymer.

We now examine the effect of transverse diffusion on the clogging rate of walls for $x > x^*$. Near the wall, there exists a layer of ideal mixing, whose length is $l^* = (0.34 \cdot DT)^{1/2}$. Outside the layer, transverse diffusion is not considered. Within the bounds of this two-layer model, the flow in the flow core can be described by Eq. (1) with the boundary conditions

$$\tau|_{x=0} = 0, \quad \tau|_{y=l^*} = T. \tag{35}$$

We introduce the variable s(x, y) such that

$$dy/ds = \varphi(x), \ dx/ds = -\gamma y;$$
 (36)

$$s\Big|_{\substack{y=l^*\\x\geqslant x^*}}=0.$$
(37)

Then (1) is transformed into the form $d\tau/ds = -1$, which from (35) gives

$$s|_{x=0} = T.$$
 (38)

Equations (36) and (37), augmented by the condition $x|_{s=0} = p$ for a given $\phi(x) > 0$, have a unique solution. Thus, it remains to find a $\phi(x)$, which fulfills condition (37) for any p. From (36) we obtain $dy/dx = -\phi(x)/(\gamma y)$, from which

$$y = \left[\frac{2}{\gamma} \int_{x}^{p} \varphi(z) \, dz + l^{*2}\right]^{1/2}.$$
(39)

Substituting (39) into (36) and considering the boundary conditions (37) and (38) yields p

$$\int_{0}^{p} \frac{dx}{\dot{\gamma} \left[\frac{2}{\dot{\gamma}} \int_{x}^{p} \varphi(z) \, dz + l^{*2}\right]^{1/2}} = T.$$
(40)

Because the growth rate of the stagnant layer of reaction products $\phi(z)$ is zero for $z < x^*$, Eq. (40) is transformed to:

$$T = \int_{0}^{\infty} \frac{dx}{\dot{\gamma} \left[\frac{2}{\dot{\gamma}} \int_{x^{*}}^{p} \varphi(z) \, dz + l^{*2}\right]^{1/2}} + \int_{x^{*}}^{p} \frac{dx}{\dot{\gamma} \left[\frac{2}{\dot{\gamma}} \int_{x^{*}}^{p} \varphi(z) \, dz + l^{*2}\right]^{1/2}} =$$
$$= \frac{x^{*}}{\dot{\gamma} \left[\frac{2}{\dot{\gamma}} \int_{x^{*}}^{p} \varphi(z) \, dz + l^{*2}\right]^{1/2}} + \int_{x^{*}}^{p} \frac{dx}{\dot{\gamma} \left[\frac{2}{\dot{\gamma}} \left(\int_{x^{*}}^{p} \varphi(z) \, dz - \int_{x^{*}}^{x} \varphi(z) \, dz\right) + l^{*2}\right]^{1/2}}$$

We make a change of variables $\hat{p} = p - x^*$, $\hat{z} = z - x^*$, and $\hat{x} = x - x^*$. Then

$$T = \frac{x^*}{l^* \dot{\gamma} [\chi(t) + 1]^{1/2}} + \int_0^p \frac{dx}{l^* \dot{\gamma} [\chi(\widehat{p}) - \chi(\widehat{x}) + 1]^{1/2}},$$
(41)

where

$$\chi(\widehat{p}) = \frac{2}{l^{*2}\gamma} \int_{0}^{p} \varphi(z) dz.$$
(42)

We introduce new variables: $\tilde{p} = \hat{p}/x^*$, $\tilde{x} = \hat{x}/x^*$, and $\tilde{\chi}(\tilde{p}) = \chi(\tilde{p}x^*)$. Then Eq. (40) takes the form



$$1 = \frac{1}{\sqrt{\tilde{\chi}(\tilde{p}) + 1}} + \int_{0}^{\tilde{p}} \frac{d\tilde{x}}{\sqrt{\tilde{\chi}(\tilde{p}) - \tilde{\chi}(\tilde{x}) + 1}},$$
(43)

After we differentiate (43), we obtain an integral-differential equation

$$\widetilde{\chi}'(\widetilde{p}) = \frac{1}{\frac{1}{2(\widetilde{\chi}(\widetilde{p})+1)^{3/2}} + \int_{0}^{\widetilde{p}} \frac{d\widetilde{x}}{2(\widetilde{\chi}(\widetilde{p})-\widetilde{\chi}(\widetilde{x})+1)^{3/2}}},$$
(44)

which can be solved numerically by a simple algorithm. In appearance, Eq. (44) resembles the Cauchy problem; however, its right side contains a definite interval of the dependent function, whose upper limit is the independent variable. Therefore, this integral must be calculated anew at each step of the numerical integration. However, sufficient accuracy can be attained in the numerical solution with a moderate volume of computations if we use the quadratic Newton-Cotes formula of high order (seventh and above) for calculating the definite integral and the Runge-Kutta method for solving the Cauchy problem.

Once the function $\chi(\tilde{p})$ is found, it is easy to use (42) to find the growth rate of the stagnant layer $\phi(\tilde{p})$ considering diffusion:

$$\varphi(\widetilde{p}) = \frac{\dot{\gamma} l^{*2}}{2} \frac{d\widetilde{p}}{d\widetilde{p}} \widetilde{\chi}'(\widetilde{p}) = \frac{l^*}{2T} \widetilde{\chi}'(\widetilde{p})$$

We remember that the growth rate ϕ_0 of the stagnant layer is determined by the rela-

tionship
$$\phi_0 = \pi^2 x/(4\dot{\gamma}T^2) = (\pi^2/4)(\ell */T)(1 + \tilde{p})$$
, from which $\frac{\varphi}{\varphi_0} = \frac{2}{\pi^2} \frac{\tilde{\chi'}(\tilde{p})}{1 + \tilde{p}}$.

From the dependence of ϕ/ϕ_0 on the dimensionless longitudinal coordinate shown in Fig. 2, it follows that transverse diffusion has a marked effect of the clogging rate in a section of length $5x^*-10x^*$, where x^* is determined from (34).

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