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CIRCULATION TIME PREDICTION IN THE SCALE-UP OF POLYMERIZATION REACTORS WITH HELICAL RIBBON AGITATORS.

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ABSTRACT

A fundamental model of fluid circulation in polymerization reactors agitated by helical ribbon impellers is presented. An expression is developed for the circulatory flow generated by a double helical impeller in a reactor with a draught tube. The product of circulation time and ribbon rotational speed is predicted to be a function only of geometric variables for Newtonian fluids in the viscous flow regime. The circulation of Newtonian fluids in an experimental reactor is investigated to verify the model. There is good agreement between the model and experimental results for Reynolds numbers less than 500. At higher Reynolds numbers, inertial effects increase fluid circulation times. Preliminary data are also presented for non-Newtonian fluids. The implications of these results in the scale-up of polymerization reactors are discussed.

KEYWORDS

Mixing, viscous fluids, non-Newtonian fluids, circulation time, polymerization reactor, helical ribbon, scale-up.

INTRODUCTION

Mixing in continuous polymerization reactors is particularly difficult because of the high viscosities and low diffusivities typically found in polymeric systems. Poor macro-mixing results in the formation of dead zones, temperature and concentration gradients, and loss of catalyst activity, leading to poor product quality and fouling. Non-Newtonian, shear-thinning effects can reduce further the size of the well-mixed zone in the reaction vessel. Helical ribbon agitators are widely used in polymerization reactors because of their ability to keep the entire vessel contents circulating. Closed streamline circulation patterns in the vessel can, however, result in dead zone formation (Takahashi *et al.*, 1982). Van Dierendonck *et al.* (1980) proposed a helical ribbon agitator with draught tube as an alternative design (Fig. 1). Propeller stirrers were provided in the draught tube to enhance micro-mixing. Little clearance between the helix and the vessel and draught tube walls ensures circulation of all fluid elements.

A key parameter in the successful application of helical impellers to continuous polymerization is the fluid circulation time. Minimizing circulation time minimizes temperature and concentration variation in the reactor by reducing the time between successive passings of the monomer feed inlet. Thus, the estimation of fluid circulation time is vital for the optimization and

scale-up of the reactor. Bourne and Butler (1969), in an investigation of circulation patterns in vessels agitated by helical impellers, proposed the following expression for the flow rate, Q :

$$Q = \frac{\pi}{2} D^2 W (\omega_b - \omega_f) \sin\theta \cos\theta \quad (1)$$

where D is the average impeller diameter, θ is the helix angle, W is the impeller width, and ω_b and ω_f are respectively the angular velocities of the impeller and the fluid well ahead of the advancing impeller. Good agreement with experimental results was obtained (average error 7%) but the expression is not fully predictive as ω_f must be empirically obtained. Carreau *et al.* (1976) proposed a similar expression for Q :

$$Q = \alpha N_b D W Z N \sin\theta \cos\theta \quad (2)$$

where N_b is the number of impeller blades, Z is the length of the impeller blades, N is the impeller speed, and α is an empirical parameter. Axial velocity profiles were obtained using tracer beads but the data were not sufficiently accurate to estimate α . Chavan and Ulbrecht (1972) used an extruder analysis to predict the pumping capacity of a helical screw agitator within a draught tube. Theoretical predictions did not agree with experimental results. Van Dierendonck *et al.* (1980) defined the maximum pumping capacity of a helical ribbon with a draught tube as the volume

of fluid displaced by the impeller in one rotation. i.e.

$$Q_{max} = N q \frac{\pi}{4} (D_O^2 - D_I^2) \quad (3)$$

where D_O and D_I are the reactor wall and draught tube diameters respectively, and q is the screw pitch. Actual pumping efficiencies were found to be about 40% of Q_{max} .

Fig. 1 Polymerization reactor with helical ribbon agitator.

MATHEMATICAL MODEL

A model of the pumping action of a double helical impeller with draught tube for low Reynolds numbers (inertial effects negligible) is developed below. The approach is based on that used in the analysis of single screw extruders (see e.g. McKelvey (1962), Middleman (1977)). The real system of two helical ribbons rotating at speed N in an annulus with stationary walls is replaced by the equivalent system of a stationary helix with the reactor and draught tube walls rotating at speed N in the opposite direction.

Fig. 3 Co-ordinate system and boundary conditions.

These moving walls drag fluid through helically spiralling channels of approximately rectangular cross-section. For a double helix there are two such channels (Fig. 2). The channel is then unwound resulting in a simpler rectangular geometry (Fig. 3). The channel dimensions are:

width, $W = \frac{\pi D \sin \theta}{2}$, is the perpendicular distance between flights; height, $H = \frac{D_O - D_I}{2}$, is the distance between the draught tube and reactor walls; and length, $Z = \frac{L}{\sin \theta}$, is the contour length of the helix. D_O , D_I , and D are the outside, inside, and average diameters of the annulus, θ is the helix angle, and L is the vertical height of the helix.

In the limit of a very narrow annulus, $\frac{H}{W} \rightarrow 0$, a one-dimensional solution to this problem can easily be obtained. i.e. Assuming:

$$\begin{aligned} v_x &= v_x(y) \\ v_z &= v_z(y) \\ v_y &= 0 \end{aligned} \quad (4)$$

Fig. 2 Unwound rectangular channel model.

the Navier-Stokes equations reduce to:

$$0 = -\frac{\partial P}{\partial x} + \mu \frac{d^2 v_x}{dy^2} \quad (5a)$$

$$0 = -\frac{\partial P}{\partial z} + \mu \frac{d^2 v_z}{dy^2} \quad (5b)$$

with boundary conditions:

$$\left. \begin{aligned} v_x &= -V \sin \theta \\ v_z &= V \cos \theta \end{aligned} \right\} y = \pm \frac{H}{2}; \quad \left. \begin{aligned} \frac{dv_x}{dy} &= 0 \\ \frac{dv_z}{dy} &= 0 \end{aligned} \right\} y = 0 \quad (6)$$

The Navier-Stokes Equations (eqns. 5a, 5b) are independent and only eqn. 5b needs to be solved to calculate the flow rate, Q . The recirculating flow in the x -direction (eqn. 5a) is, however, important in estimating residence time distribution and total viscous shear mixing in the helix. Integration of eqn. 5b with respect y yields the following expression for the velocity along the channel, v_z :

$$v_z = V \cos \theta - \frac{\Delta P_H}{2\mu Z} \left[\left(\frac{H}{2} \right)^2 - y^2 \right] \quad (7)$$

where $\frac{\partial P}{\partial z}$ has been replaced by $\frac{\Delta P_H}{Z}$ where ΔP_H is the total pressure drop across the helix. The flow rate, Q , through the helix (2 channels) is then obtained by integration of eqn. 7. i.e.

$$Q = 2W \int_{-\frac{H}{2}}^{\frac{H}{2}} v_z dy = 2WVH \cos \theta - \frac{WH}{6Z} \frac{\Delta P_H}{\mu} \quad (8)$$

Substituting $V = N\pi D$, $L = Z \sin \theta$, and $W = \frac{\pi D \sin \theta}{2}$ gives:

$$Q = (\pi^2 D^2 H \sin \theta \cos \theta) N - \frac{\pi D H^3 \sin^2 \theta}{12L} \left(\frac{\Delta P_H}{\mu} \right) \quad (9)$$

The assumption that the aspect ratio $\frac{H}{W} \ll 1$ is generally not valid for helical impellers in a draught tube ($\frac{H}{W} \approx O(1)$). Thus, in place of the 1-D expression ($v_z = v_z(y)$, eqn. 5b), the 2-D, z -direction Navier-Stokes Equation must be solved ($v_z = v_z(x, y)$). i.e.

$$0 = -\frac{\partial P}{\partial z} + \mu \left[\frac{\partial^2 v_z}{\partial x^2} + \frac{\partial^2 v_z}{\partial y^2} \right] \quad (10)$$

with boundary conditions:

$$\left. \begin{aligned} v_z &= V \cos \theta, & y &= \pm \frac{H}{2} \\ v_z &= 0, & x &= \pm \frac{W}{2} \end{aligned} \right\} \quad (11)$$

The method of separation of variables is applied to solve this boundary value problem (see e.g. McKelvey). Physically the drag flow induced by the moving walls and the pressure driven flow can be obtained separately and summed. The following expression is obtained for the flow rate

through the helix:

$$Q = (\pi^2 D^2 H F_D \sin \theta \cos \theta) N - \frac{\pi D H^3 F_P \sin^2 \theta}{12L} \left(\frac{\Delta P_H}{\mu} \right) \quad (12)$$

Thus, the 2-D solution, eqn. 12, has the same form as the 1-D solution, eqn. 9, but with the addition of shape correction factors, F_D and F_P . These are the following functions of the aspect ratio of the rectangular channel, $\frac{H}{W}$ (McKelvey):

$$F_D = \frac{16W}{\pi^3 H} \sum_{i=1,3,\dots}^{\infty} \frac{1}{i^3} \tanh \left[\frac{i\pi H}{2W} \right] \quad (13a)$$

$$F_P = 1 - \frac{192H}{\pi^5 W} \sum_{i=1,3,\dots}^{\infty} \frac{1}{i^5} \tanh \left[\frac{i\pi W}{2H} \right] \quad (13b)$$

F_D and F_P are plotted as a function of $\frac{H}{W}$ in Figure 4. Equation 12 is of the form

$$Q = AN - C \left(\frac{\Delta P_H}{\mu} \right) \quad (14)$$

where $A = (\pi^2 D^2 H F_D \sin \theta \cos \theta)$ and

$C = \frac{\pi D H^3 F_P \sin^2 \theta}{12L}$ are functions only of reactor geometry.

Equation 14 still contains one unknown: the pressure gradient across the helix, ΔP_H . This must equal the pressure drop across the rest of the reactor. i.e. $\Delta P_H = \Delta P_{E,1} + \Delta P_D + \Delta P_{E,2}$ where ΔP_D is the pressure drop along the draught tube. Flow through the draught tube (containing a stationary propeller shaft) can be modelled as Poiseuille flow in a cylindrical annulus. The solution can be written as (Bird *et al.*):

$$Q = k \left(\frac{\Delta P_D}{\mu} \right) \quad (15)$$

where $k = \frac{\pi R_D^4}{8L} \left[\left(1 - \lambda^4 \right) - \frac{(1 - \lambda^2)^2}{1n(1/\lambda)} \right]$; λ is the

ratio of shaft to draught tube diameters: $\lambda = \frac{R_S}{R_D}$. Neglecting the end pressure drops, $\Delta P_{E,1}$ and $\Delta P_{E,2}$, implies $\Delta P_H = \Delta P_D$ and eqns. 14 and 15 can be solved to eliminate pressure:

$$Q = \frac{A}{C + k} N \quad (16)$$

The fluid circulation time, t_c , can then be calculated.

$$t_c = \frac{V_R}{Q} = \frac{C + k}{Ak} \frac{V_R}{N} \quad (17)$$

$$\text{or} \quad Nt_c = \frac{V_R}{A} \left[1 + \frac{C}{k} \right] \quad (18)$$

where V_R is the reactor volume. Thus the product Nt_c is predicted to be a function only of geometric variables and independent of fluid viscosity.

Fig. 4 Rectangular channel shape factors.

In this analysis the pumping effect of the propeller stirrers is neglected. The primary function of stirrers in the draught tube is to enhance micro-mixing and radial dispersion of fluid elements. The propeller stirrers will cause a increase in circulatory flow rate, Q , and thus decrease t_c . However at commercial operating conditions this effect is small.

EXPERIMENTAL APPARATUS

A model reactor with a plexiglas wall and draught tube were constructed to investigate fluid circulation and mixing. The reactor dimensions were $V_R=0.166\text{m}^3$, $D_O=0.477\text{m}$ and $H=0.0985\text{m}$. A double helix of angle $\theta=13.6^\circ$ was used in the experiments. The draught tube contained a propeller stirrer which was kept stationary for the experiments reported here. Applying eqns. 14 and 15: $A=0.21\text{m}^3$, $C=5.4\times 10^{-6}\text{m}^3$, $k=1.5\times 10^{-4}\text{m}^3$, and thus $Nt_c=8.2$.

The iodine/sodium thiosulphate technique was used to experimentally investigate fluid circulation in the model reactor. Mixtures of glucose syrup and water of varying viscosity were used as experimental Newtonian fluids. Approximately 5mls of 1M I_2 dissolved in a glucose syrup/water solution of the same viscosity as the test fluid was injected into the reactor. The circulation of tracer was monitored at two points using light sensitive detectors (Figure 5). A typical detector response is shown in Figure 6. The average period of the output signal is the circulation time of fluid in the reactor. 1M sodium thiosulphate was used to decolourize the fluid before the next injection.

RESULTS

Newtonian Fluids

Circulation time data were obtained with 11 Newtonian fluids of viscosity ranging from $23\text{Pa}\cdot\text{s}$ (pure glucose syrup) to $0.001\text{Pa}\cdot\text{s}$ (pure water). Results obtained with the helix pumping upwards and downwards are shown in Figure 7. The results are plotted as the product of helix rotation speed and circulation time, Nt_c , as a function of

Fig. 5 Tracer injection and detection positions.

Reynolds number, Re .

$$Re = \frac{\rho N D_O^2}{\mu} \quad (19)$$

Nt_c is independent of Re and pumping direction for $Re < 500$. There is good agreement between low Reynolds number Nt_c , $Nt_c = 9.5 \pm 0.5$, and the prediction of the creeping flow model, $Nt_c = 8.2$. Differences between the two are due to complexities of the reactor not accounted for in the model such as: helix support shafts, leakage flows between helix and walls, channel curvature, and the flow resistance of the top and bottom section. Above a critical Reynolds number, $Re_{ct}=500$, Nt_c increases with Re because of inertial effects. Tube entrance and exit pressure drops increase with Re (Boger, 1982). There is also a separation of upwards and downwards pumping because of the inertial pumping action of the helix support (at the bottom of the reactor, Fig. 1). The helix support acts as a centrifugal pump and will always pump outwards (and thus upwards in the helix annulus) regardless of the helix pumping direction. Thus it will enhance fluid circulation for an upward pumping helix and conversely retard circulation for a downward pumping helix. The value of Re_{ct} below which Nt_c is constant is dependent on the detailed design of helix supports and other reactor internals.

Non-Newtonian Fluids

Circulation experiments were also performed with three non-Newtonian fluids: aqueous CMC solutions. The rheological properties of the fluids are plotted in Figure 8. The viscosity of these highly shear-thinning fluids can be modelled by a power law equation:

$$\tau = K \dot{\gamma}^n \quad (20)$$

where τ is the shear stress and $\dot{\gamma}$ the shear rate. Parameters in a power law fit of the viscosity vs. shear rate data for the CMC solutions are given in Table 1. A generalized Reynolds number for shear-thinning fluids, Re^* , can be defined (Metzner and Otto, 1957):

$$Re^* = \frac{\rho N D_O^2}{\mu_{ef}} \quad (21)$$

Fig. 6 Typical detector output trace showing circulation and mixing times.

Fig. 7 Circulation data for Newtonian fluids.

Fig. 8 Viscosity vs. shear rate for CMC solutions.

Fig. 9 Circulation data for non-Newtonian fluids.

Table 1
Power law parameters for CMC solutions

C (wt. %)	K Pa.s n	n
0.8	0.183	0.75
1.3	1.30	0.67
2.0	6.0	0.61

where μ_{ef} is the effective viscosity. Selecting as a velocity scale the linear velocity of the helix: πND ; and as a length scale half the channel width: $H/2$; the effective shear rate inside the helical channels is given as $\gamma_{ef} = 2\pi ND/H$ (i.e. for the helical impeller of this work $\gamma_{ef} = 25$) and the effective viscosity is given by:

$$\mu_{ef} = K \left[\frac{2\pi ND}{H} \right]^{n-1} \quad (22)$$

The experimental circulation time data for CMC solutions are plotted together with the Newtonian fluid data in Fig. 9. Nt_c values for these moderately shear-thinning fluids ($0.61 < n < 0.75$) are higher than for Newtonian fluids. At lower Reynolds numbers, the difference ($< 30\%$) is not great and the model can still provide a rough estimate of t_c . However, the divergence of upwards and downwards pumping is greater and occurs at a lower Re than in the Newtonian case. Thus great care must be exercised in applying high Reynolds number, Newtonian results to the shear-thinning polymer solutions often found in polymerization reactors. Extension of the viscous flow model to power law fluids is currently being investigated.

SCALE-UP

In the viscous flow regime ($Re < Re_{ct}$), eqn. 18 provides a quantitative prediction of the effect of various geometric parameters (D_O, H, θ, V_R) on Nt_c .

The term $\frac{C}{k}$ is usually small. Thus:

$$Nt_c \approx \frac{V_R}{A} = \frac{V_R}{\pi^2 D^2 H F_D \sin \theta \cos \theta} \quad (23)$$

Since both V_R and A scale with D^3 ($H \propto D$), Nt_c is independent of scale for geometrically similar reactors. Operation at the same rotational speed, N , is sufficient to ensure no change in t_c when scaling up a reactor.

In scaling up from a pilot plant to a commercial polymerization reactor, typically the reaction fluid properties are unchanged. Scale-up using the same fluid (ρ and μ constant) implies an increase in Re which may then exceed the critical value. Thus, while the pilot plant may be in the viscous flow regime, the full-scale commercial plant may be in an inertially dominated regime ($Re > Re_{ct}$). Circulation time can greatly increase (Fig. 7) and will be a function of pumping direction and reactor internal details. The size and shape of the helix support and other internals play an important role in the inertial regime. Thus, the mechanical limitations imposed on the full-scale reactor should be considered in pilot plant design. The pilot plant reactor should be scaled down from the commercial reactor.

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NOMENCLATURE

A	drag flow geometric constant	m^3
C	pressure flow geometric constant	m^3
D_O	impeller outside diameter	m
D_I	impeller inside diameter	m

D	average impeller diameter	m
H	annulus width	m
k	draught tube geometric constant	m^3
K	power law consistency factor	$Pa.s^n$
L	height of helical impeller	m
n	power law index	
N	rotational speed of impeller	s^{-1}
q	helix pitch	m
Q	flow rate	m^3/s
R_D	draught tube inside radius	m
R_P	propeller shaft diameter	m
Re	Reynolds number	
Re^*	generalized Reynolds number	
Re_{ct}	critical Reynolds number	
t_c	circulation time	s
t_m	mixing time	s
V_R	reactor volume	m^3
V	impeller velocity at diameter D	m/s
v_i	velocity in i -direction ($i = x, y, z$)	m/s
W	perpendicular distance between helix flights	m
Z	length of flow path through helix	m
x, y, z	co-ordinate directions	m
γ	shear rate	s^{-1}
γ_{ef}	effective shear rate	s^{-1}
ΔP_H	pressure change across helix	Pa
ΔP_D	pressure drop across draught tube	Pa
η	non-Newtonian fluid viscosity	$Pa.s$
λ	shaft to draught tube radius ratio	
μ	Newtonian fluid viscosity	$Pa.s$
μ_{ef}	effective non-Newtonian viscosity	$Pa.s$
ρ	density	kg/m^3
τ	shear stress	Pa

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