

Simulation of Nylon 6 Polymerization in Tubular Reactors with Recycle

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Synopsis

In the hydrolytic polymerization of ϵ -caprolactam, the ring opening of the monomer is much slower than the polyaddition reaction. Hence, the mixing of aminocaproic acid to the feed results in a faster conversion of the monomer. Industrially, this fact is exploited by using a recycle stream. An isothermal plug flow reactor (PFR) with a recycle is simulated in this study, using two techniques: the method of successive substitutions and Wegstein's method. It is found that, under certain operating conditions, the use of a recycle stream gives higher monomer conversions and lower cyclic dimer concentrations than either a PFR or a homogeneous continuous-flow stirred-tank reactor (HCSTR), with the degree of polymerization almost the same as that obtained in an HCSTR, and thus offers a considerable advantage. However, when a recycle reactor is coupled with a subsequent flashing operation and a finishing reactor, these advantages are considerably reduced.

INTRODUCTION

The simulation and optimization of industrial nylon 6 reactors has been the subject of considerable study in recent years.¹⁻¹⁷ The polymerization of ϵ -caprolactam has been modeled in terms of three parallel reactions: ring opening of ϵ -caprolactam by water, polyaddition occurring between polymer molecules and ϵ -caprolactam, and polycondensation between polymer molecules. In addition to these major reactions, several side reactions also occur which influence the quality of the final polymer appreciably. These have been reviewed by Reimschuessel.¹ One of these is the cyclization of the linear molecules. The cyclic compounds so formed create problems in the spinning process and must be removed by an energy-intensive hot-water-extraction or a vacuum-evaporation process.

Reimschuessel and Nagasubramanian² first obtained expressions for the rate and equilibrium constants for the three major reactions from their experimental data at different temperatures. Their results were consistent with earlier experimental studies at 220°C of Hermans et al.,¹⁸ Kruissink et al.,¹⁹ and Wiloth.²⁰ These rate constants have been used by several workers²⁻⁸ to simulate isothermal as well as nonisothermal polymerizations of nylon 6 in plug-flow reactors (PFRs), homogeneous continuous-flow stirred-tank reactors (HCSTRs), and in cascades of HCSTRs. The effect of removing water instantaneously from the reaction mass after some conversion was attained, was also studied.^{2,8} Three parameters of interest, namely, the conversion of caprolactam, the number-average chain length, and the concentration of the acid end groups were computed for various process conditions, by solving appropriate mass-balance equations.

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Tirrell et al.³ extended the analysis of Reimschuessel and Nagasubramanian² and obtained the second moment of the polymer chain-length-distribution. They also incorporated the effect of adding monofunctional stabilizers to the reaction mass. In a recent analysis^{4,5} the mass-balance equations for the individual species were integrated numerically to give the entire molecular weight distribution (MWD) for isothermal batch reactors. The number-average chain length computed from the MWDs agreed well with those obtained by earlier workers^{2,3} using the functional-group analysis. Tai et al.⁹⁻¹³ have reported more extensive experimental work on the hydrolytic polymerization of ϵ -caprolactam and have curve-fitted their data using nonlinear regression to give more accurate expressions for the rate and equilibrium constants. In addition to the three major reactions, they also incorporated the reaction for the formation of the cyclic dimer and the reaction between the cyclic dimer and the linear oligomers. They omitted the reactions for the higher cyclic oligomers since the concentrations of the latter were usually much lower.

The nonisothermal polymerization of nylon 6 in VK (Vereinfacht Kontinuierliches Rohr) columns used commonly in industry has been studied by Mochuzuki and Ito,^{14,15} Jacobs and Schweigman,⁶ and Naudin den Cate,⁷ and optimal temperature and water-concentration profiles have been obtained^{7,15} using different objective functions, mathematical techniques, and kinetic schemes. Attempts have also been made to model the finite rates of evaporation of water from the reaction mass, both by the use of vacuum²¹ as well as by using a cocurrent²² flow of inert-gas bubbles. Optimization studies using these models are under progress.

Several methods to speed up the conversion of the monomer have been studied. One of these, suggested by Wiloth,²³⁻²⁵ is the use of amino-caproic acid in the feed to the reactor. This is based upon the fact that the polyaddition reaction is much faster than the ring-opening step. One method for exploiting this on an industrial scale is to recycle part of the reaction mass in a PFR, as shown in Figure 1. Such schemes have been suggested and tried in industry.^{1,26-28} In this paper, we have modeled one such recycle reactor for nylon 6 polymerization and have studied the effects of various operating variables on the product. It may be added that the improvement in the product distributions using recycle

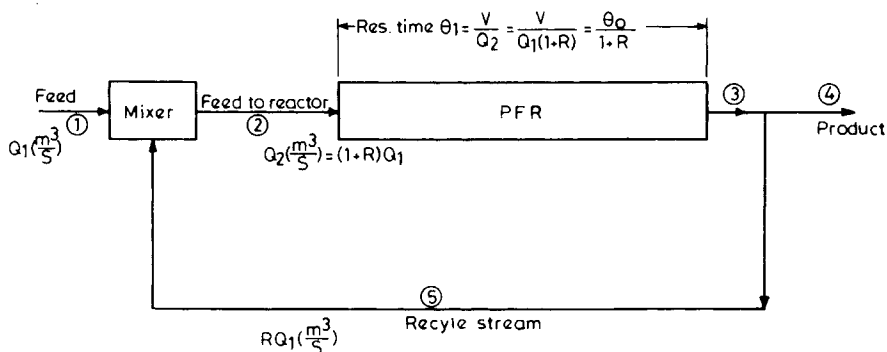


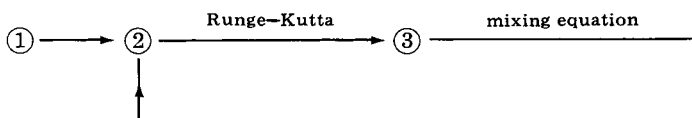
Fig. 1. Reactor configuration studied. V = volume of PFR, Q_1 = volumetric flow rate at point 1, and R = recycle ratio.

streams for low molecular weight, nonpolymeric compounds has been a subject of considerable research activity recently.²⁹

FORMULATION

The kinetic scheme used in this work (Table I) is an extension of that suggested by Tai et al.¹¹ in that it incorporates the reactions with monofunctional acids as well. Mass balance equations can be written for each molecular species using this kinetic scheme. These can be summed up appropriately to give the equations for the various moments μ_k ($\equiv \sum_{n=1}^{\infty} n^k [s_n]$, $k = 0, 1, 2, \dots$) and μ'_k ($\equiv \sum_{n=1}^{\infty} n^k [A_n]$, $k = 0, 1, 2, \dots$) of the MWDs of the bifunctional and monofunctional compounds, respectively. The final equations for the isothermal PFR are similar to those presented earlier²² and are given in Table II, along with appropriate closure equations^{12,22} for the third moments (μ_3, μ'_3) and the concentrations of S_2 and S_3 . The equations given in this table present a *complete* set of coupled, nonlinear, ordinary differential equations and can be integrated numerically using the Runge-Kutta algorithm of order 4, to give the values of the 11 variables $\mu_0, \mu'_0, \mu_1, \mu'_1, \mu_2, \mu'_2, [C_1], [C_2], [S_1], [A_1]$, and $[W]$, at the outlet of the PFR (point 3 in Fig. 1) in terms of the corresponding values at the feed to the reactor (point 2), for a given value of the residence time, θ_1 . However, because of the recycling, the conditions at point 2 depend not only on the values at point 1, which are known, but also on the values at point 3, the corresponding mixing equations being given in Table II. The solution of this set of 22 equations (11 for the reactor and 11 for the mixer) thus requires a method of arriving at the right set of values of these variables at point 2. When these are used with the Runge-Kutta algorithm to compute the conditions at point 3, and, thereafter, when these are used in the mixing equations to recompute the conditions at point 2, the same results should be obtained.

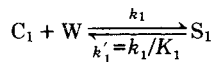
Several techniques have been tried for nonpolymeric reactors for attaining this convergence, of which two techniques are commonly used.³⁰ In the first technique, called the method of successive substitutions, one keeps traveling in the following computational cycle, till computed results at point 2 are identical



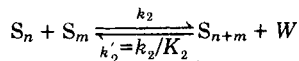
to that in the previous cycle, within the limits of convergence. This technique takes a relatively longer time to converge compared to the second technique of Wegstein,³⁰ which speeds up or retards the approach towards the solution. The major difference between this method and that of successive substitutions is that at any stage, once the conditions at point 2 are known from the mixing equations, instead of using these as the feed to the reactor, a different "educated" guess (computed from the previous conditions at point 2) is used which extrapolates and accelerates the path towards convergence. The degree of acceleration can, however, be controlled by setting limits on the values of the "acceleration parameters,"³⁰ t_i . Evidently, the faster the desired convergence, the more the likelihood of this method oscillating and diverging.

TABLE I
Kinetic Scheme and Data¹² for Nylon-6 Polymerization^a

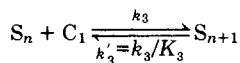
1. Ring opening



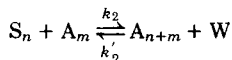
2. Polycondensation



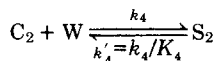
3. Polyaddition



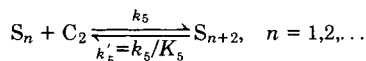
4. Reaction with monofunctional acid



5. Ring opening of cyclic dimer



6. Polyaddition of cyclic dimer



Rate and Equilibrium Constants

$$k_i = A_i^0 \exp\left(-\frac{E_i^0}{RT}\right) + A_i^c \exp\left(-\frac{E_i^c}{RT}\right) \sum_{n=1}^{\infty} [S_n]$$

$$K_i = \exp\left(\frac{\Delta S_i - \Delta H_i/T}{R}\right), \quad i = 1, 2, \dots, 5$$

<i>i</i>	A_i^0 (kg/mol·h)	E_i^0 (cal/mol)	A_i^c (kg ² /mol ² ·h)	E_i^c (cal/mol)	ΔH_i (cal/mol)	ΔS_i (e.u.)
1	5.9874×10^5	1.9880×10^4	4.3075×10^7	1.8806×10^4	1.9180×10^3	-7.8846×10^0
2	1.8942×10^{10}	2.3271×10^4	1.2114×10^{10}	2.0670×10^4	-5.9458×10^3	9.4374×10^{-1}
3	2.8558×10^9	2.2845×10^4	1.6377×10^{10}	2.0107×10^4	-4.0438×10^3	-6.9457×10^0
4	8.5778×10^{11}	4.2000×10^4	2.3307×10^{12}	3.7400×10^4	-9.6000×10^3	-1.4520×10^1
5	2.5701×10^8	2.1300×10^4	3.0110×10^9	2.0400×10^4	-3.1691×10^3	5.8265×10^{-1}

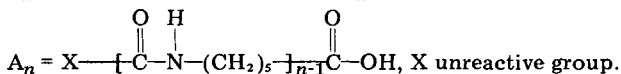
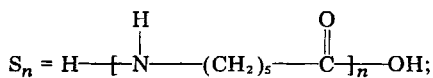
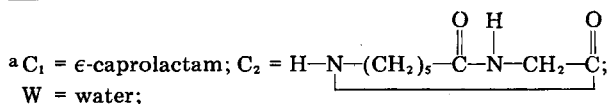


TABLE II
Mass Balance Equations For the Isothermal PFR: $0 \leq t \leq \theta_1$

$$\frac{\partial \mu_0}{\partial t} = k_1[C_1][W] - k'_1[S_1] - k_2\mu_0^2 + k'_2[W](\mu_1 - \mu_0) - k_2\mu_0\mu'_0 + k'_2[W](\mu'_1 - \mu'_0) + k_4[W][C_2] - k'_4[S_2]$$

$$\frac{\partial \mu'_0}{\partial t} = 0$$

$$\begin{aligned} \frac{\partial \mu_1}{\partial t} = & k_1[C_1][W] - k'_1[S_1] + k_3[C_1]\mu_0 - k'_3(\mu_0 - [S_1]) \\ & - k_2\mu'_2\mu_1 - k'_2[W]\left(\frac{\mu'_1 - \mu'_2}{2}\right) + 2k_5[C_2]\mu_0 \\ & - 2k'_5(\mu_0 - [S_1] - [S_2]) + 2k_4[W][C_2] - 2k'_4[S_2] \end{aligned}$$

$$\frac{\partial \mu'_1}{\partial t} = k_2\mu_1\mu'_0 - \frac{k'_2[W]}{2}(\mu'_2 - \mu'_1)$$

$$\begin{aligned} \frac{\partial \mu_2}{\partial t} = & k_1[C_1][W] - k'_1[S_1] + 2k_2\mu_1^2 + \frac{k'_2}{3}[W](\mu_1 - \mu_3) \\ & + k_3[C_1](\mu_0 + 2\mu_1) + k'_3(\mu_0 - 2\mu_1 + [S_1]) - k_2\mu_2\mu'_0 + \frac{k'_2}{6}[W]*(2\mu'_3 - 3\mu'_2 + \mu'_1) \\ & + 4k_5[C_2](\mu_1 + \mu_0) + 4k'_5(\mu_0 - \mu_1 + [S_2]) + 4k_4[W][C_2] - 4k'_4[S_2] \end{aligned}$$

$$\frac{\partial \mu'_2}{\partial t} = k_2(2\mu_1\mu'_1 + \mu_2\mu'_0) - \frac{k'_2[W]}{6}(4\mu'_3 - 3\mu'_2 - \mu'_1)$$

$$\frac{\partial [C_1]}{\partial t} = -k_1[C_1][W] + k'_1[S_1] - k_3[C_1]\mu_0 + k'_3(\mu_0 - [S_1])$$

$$\begin{aligned} \frac{\partial [S_1]}{\partial t} = & k_1[C_1][W] - k'_1[S_1] - 2k_2[S_1]\mu_0 + 2k'_2[W](\mu_0 - [S_1]) \\ & - k_3[S_1][C_1] + k'_3[S_2] - k_2\mu'_0[S_1] + k'_2[W](\mu'_0 - [A_1]) - k_5[S_1][C_2] + k'_5[S_3] \end{aligned}$$

$$\frac{\partial [A_1]}{\partial t} = -k_2[A_1]\mu_0 + k'_2[W](\mu'_0 - [A_1])$$

$$\frac{\partial [C_2]}{\partial t} = -k_4[C_2][W] + k'_4[S_2] - k_5[C_2]\mu_0 + k'_5(\mu_0 - [S_1] - [S_2])$$

$$\begin{aligned} \frac{\partial [W]}{\partial t} = & -k_1[C_1][W] + k'_1[S_1] + k_2(\mu_0)^2 - k'_2[W](\mu_1 - \mu_0) \\ & + k_2\mu_0\mu'_0 - k'_2[W](\mu'_1 - \mu'_0) - k_4[C_2][W] + k'_4[S_2] \end{aligned}$$

Closure conditions: $[S_2] = [S_3] = [S_1]$

$$\mu_3 = \frac{\mu_2(2\mu_2\mu_0 - \mu_1^2)}{\mu_1\mu_0}, \quad \mu'_3 = \frac{\mu'_2(2\mu'_2\mu'_0 - \mu_1'^2)}{\mu'_1\mu'_0}$$

Initial Conditions: at $t = 0$, feed conditions same as output from mixer
For the Mixer (Fig. 1)

$$x_{\textcircled{2}} = [1/(1 + R)](x_{\textcircled{1}} + Rx_{\textcircled{5}}) = [1/(1 + R)](x_{\textcircled{1}} + Rx_{\textcircled{3}})$$

points ①, ②, ③, and ⑤ as in Figure 1

x can be μ_0 , μ'_0 , μ_1 , μ'_1 , μ_2 , μ'_2 , $[C_1]$, $[S_1]$, $[A_1]$, $[C_2]$, or $[W]$

Several checks were made to ensure that the program was free of errors. The following overall balance equation for the concentration of $(\text{CH}_2)_5$ repeat units was tested

$$[A_1]_1 + [C_1]_1 = \left\{ [C_1] + 2[C_2] + \sum_{n=1}^{\infty} (n[S_n] + n[A_n]) \right\}_3 \quad (1)$$

where $[A_1]_i, \dots$ represent the concentrations of A_1, \dots at point i of Figure 1. Results were found to lie within about 0.005%. Similarly, the following overall balance on the water concentration was checked,

$$[W]_1 = \left([W] + \sum_{n=1}^{\infty} [S_n] \right)_3 \quad (2)$$

and, again, results were found to satisfy this equation within about 0.01%. In addition, the reaction got quenched when no water was used in the feed stream (point 1), consistent with eq. (2). It was also found that, for some cases for which convergence was attained, the same final results were obtained from Wegstein's method as from the successive substitution technique, but, interestingly, both techniques required the same number of iterations. However, with some values of the recycle ratio R , Wegstein's technique failed to converge, even when limits were placed on the acceleration parameters t_i . No similar convergence problems were faced with the method of successive substitutions over the entire range of variables studied. It may be added that Wegstein's method becomes identical to the method of successive substitutions when the t_i 's are independently chosen as unity.

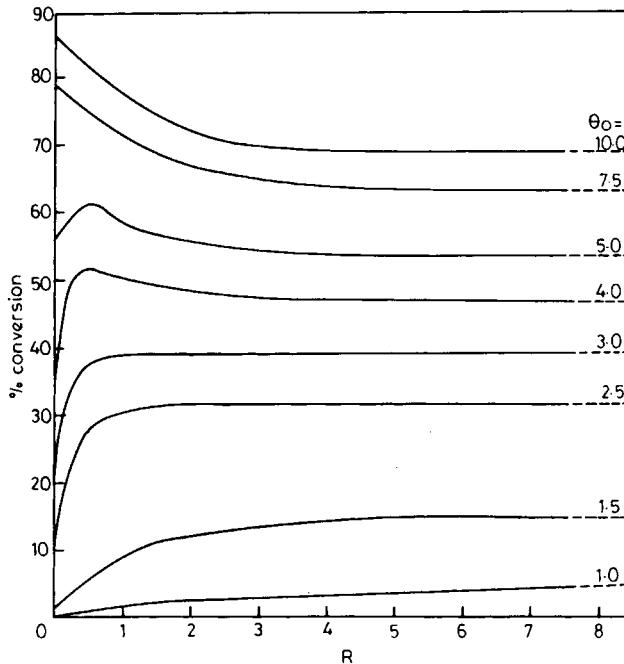


Fig. 2. Caprolactam conversion at point 3 for different R and θ_0 (defined in Fig. 1). $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$. Dotted lines represent HCSTR results for the same feed.

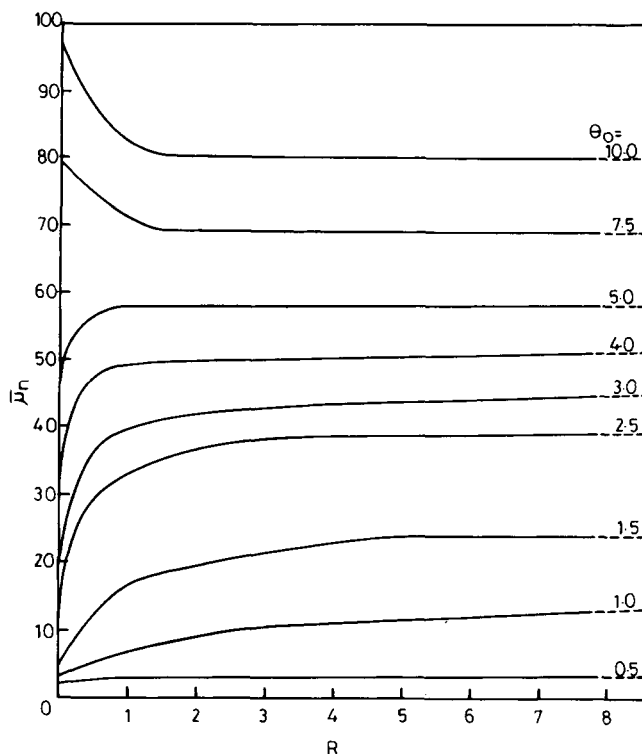


Fig. 3. $\bar{\mu}_n$ at point 3 for different R and θ_0 (defined in Fig. 1). $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$. Dotted lines represent HCSTR results for the same feed.

RESULTS AND DISCUSSION

Several simulations were carried out to study the effect of various parameters. Figures 2–4 show the effect of the recycle ratio R defined in Figure 1, keeping

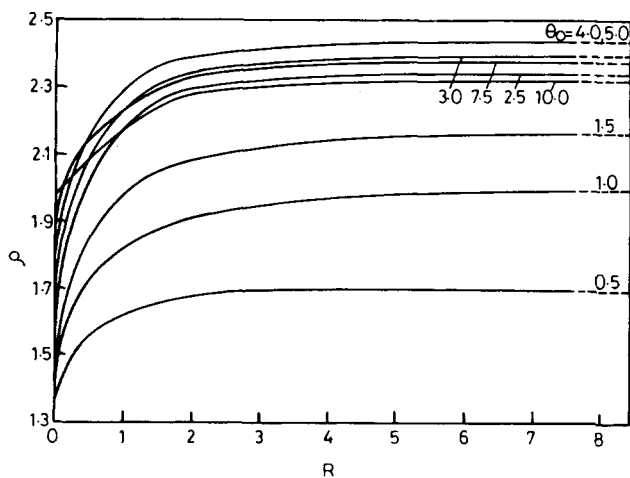


Fig. 4. ρ at point 3 for different R and θ_0 (defined in Fig. 1). $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$. Dotted lines represent HCSTR results for the same feed.

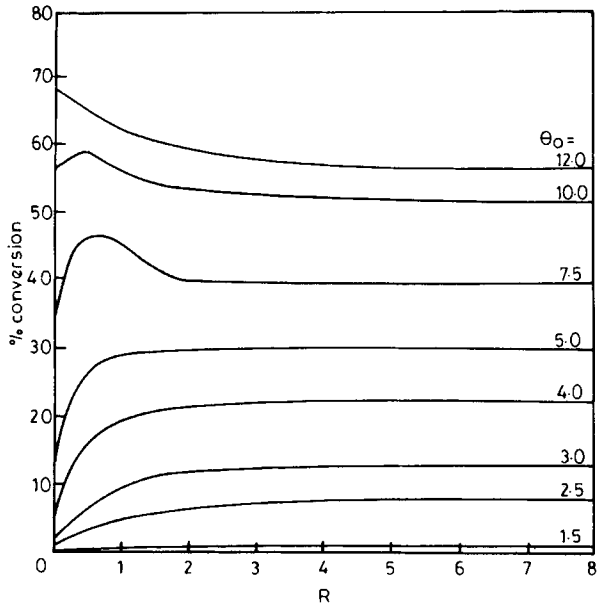


Fig. 5. Caprolactam conversion at point 3 for different R and θ_0 . $[W]_1 = 0.22$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$.

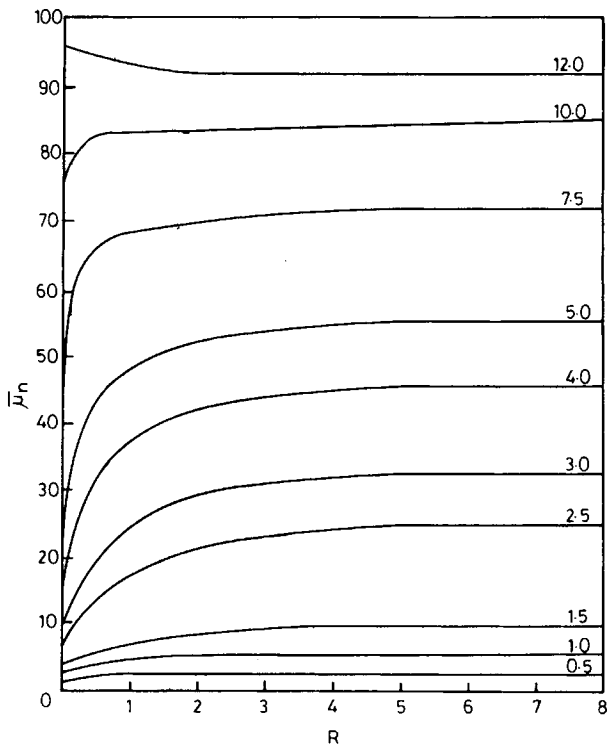


Fig. 6. $\bar{\mu}_n$ at point 3 for different R and θ_0 . $[W]_1 = 0.22$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$.

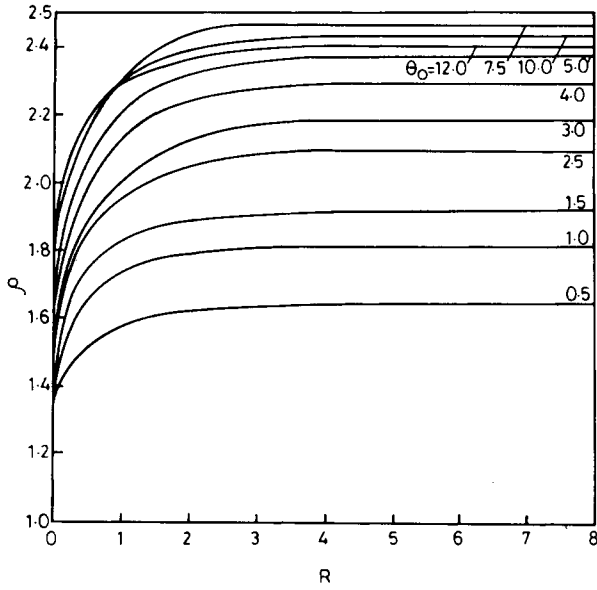


Fig. 7. ρ at point 3 for different R and θ_0 . $[W]_1 = 0.22$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 235^\circ\text{C}$.

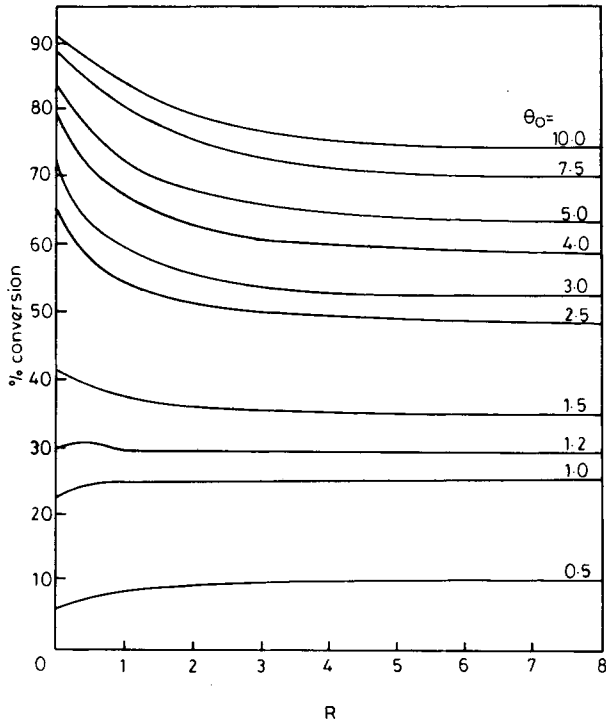


Fig. 8. Caprolactam conversion at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0.088$ mol/kg, $T = 235^\circ\text{C}$.

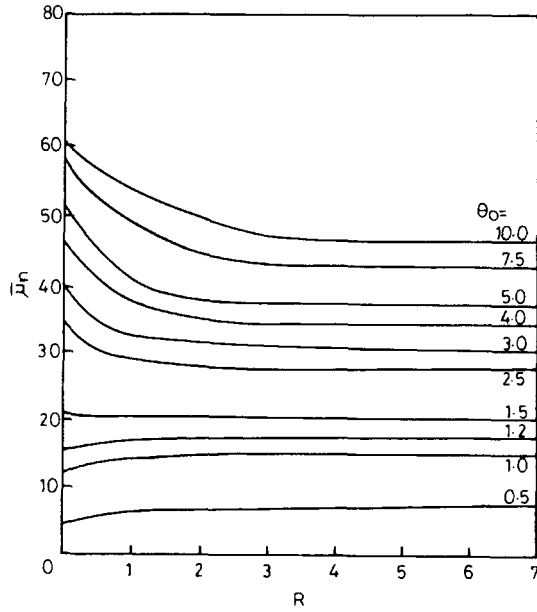


Fig. 9. $\bar{\mu}_n$ at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0.088$ mol/kg, $T = 235^\circ\text{C}$.

the volume V of the PFR constant [this means that the residence time θ_1 in Figure 1 decreases as R increases, the exact relationship between θ_1 and the flow rate at point 1, Q_1 , being given by $\theta_1 = V/Q_1(1 + R) = \theta_0/(1 + R)$]. The total computer time required to generate the results plotted in these graphs was about 20 min. It is observed for the feed conditions taken $\{[C_1]_1 = 8.8$ mol/kg, $[W]_1 = 0.44$

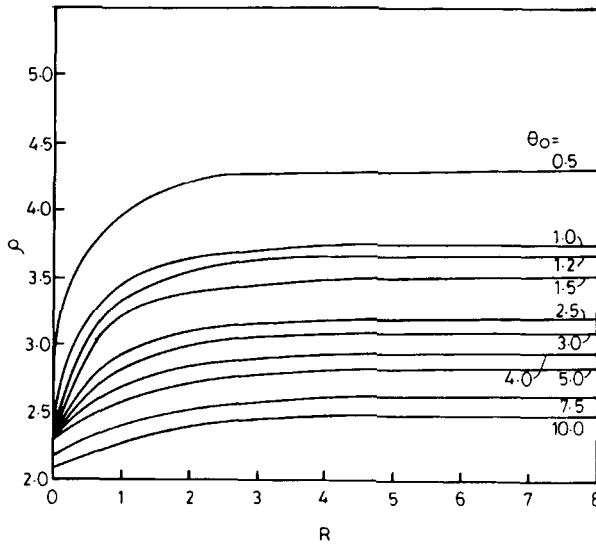


Fig. 10. ρ at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0.088$ mol/kg, $T = 235^\circ\text{C}$.

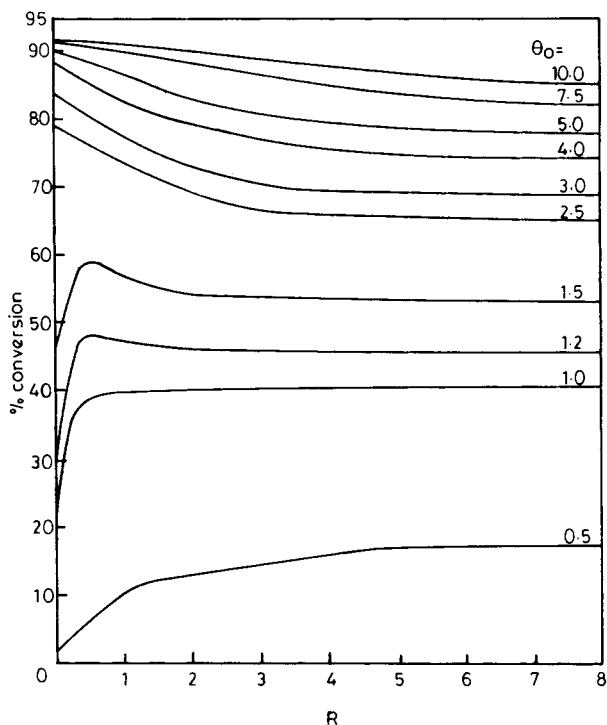


Fig. 11. Caprolactam conversion at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 265^\circ\text{C}$.

mol/kg, $T = 235^\circ\text{C}$) that as $R \rightarrow \infty$, the results for the PFR with recycle approach that for an HCSTR¹⁶ with identical feed conditions. This is expected since an HCSTR is a PFR with an infinite amount of backmixing.³¹

In Figure 2, the effect of increasing the amount of recycle at constant values of θ_0 is studied. This means that the volume of the reactor and the flow rate of the feed to the system (i.e., at point 1 of Fig. 1) is kept constant as the recycle ratio is increased. It is seen that increasing R leads to higher conversions of the caprolactam. Since $R \rightarrow \infty$ represents an HCSTR, while $R = 0$ represents an ideal PFR, this also signifies that an HCSTR gives a higher conversion of caprolactam than a PFR. This is in marked contrast to the behavior observed for nonpolymeric systems where the HCSTR leads to lower conversions, and is because of the fact that the ring-opening reaction is much slower than the polyaddition reaction.

Figure 2 shows similarly that the caprolactam conversion at $\theta_0 = 4$ h is higher when $R \rightarrow \infty$ (HCSTR) than when it is zero (PFR). However, a maxima in the conversion is observed at values of R of about 0.5. The higher conversion at intermediate values of R is once again because of the fact that the ring-opening step is much slower than the polyaddition reaction and the use of a recycle stream leads to the addition of amino groups to the feed. Furthermore, it is seen from Figure 3 that the higher value of the monomer conversion in the presence of recycle is not associated with any reduction in the number-average chain length, $\bar{\mu}_n$, of the polymer formed. Thus, there is a definite advantage in using a PFR

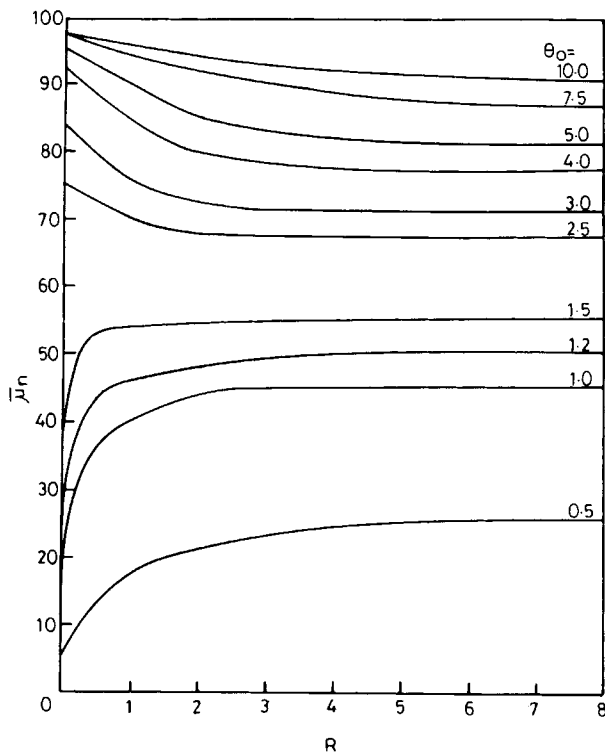


Fig. 12. $\bar{\mu}_n$ at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 265^\circ\text{C}$.

with a recycle stream under certain conditions over the use of either a PFR or an HCSTR. The polydispersity index ρ of the polymer as $R \rightarrow \infty$ for these values of θ_0 is higher than that from a recycle reactor as seen from Figure 4. This means that the product from an HCSTR will have a higher weight-average chain length than that from either a recycle reactor or a PFR. At still higher values of θ_0 , i.e., when the recycle stream is taken from a point far downstream of the feed end, it is observed from Figures 2 and 3 that a PFR without a recycle gives both the highest conversion as well as $\bar{\mu}_n$ and the use of a recycle does not offer any particular advantage. However, the recycle reactor is a good model for *real* tubular reactors and any discrepancies between computed results from a PFR and actual values from polymerization reactors may be explained easily using this model. It may be noted that slightly lower asymptotic values of ρ are obtained (Fig. 4) as θ_0 is increased.

Figures 5–7 show that similar results are obtained when a lower water concentration is used in the feed (point 1) stream. Again, the use of a recycle is found to be better than both a PFR and an HCSTR for certain values of θ_0 . It is observed that the peak in the monomer conversion occurs at higher values of θ_0 than in Figure 2. This is consistent with our earlier observation that the maxima in the concentration of S_1 are achieved farther downstream in a PFR when lower feed concentrations of water are used.²²

Figures 8–10 show the effect of using monofunctional acids in the feed stream.

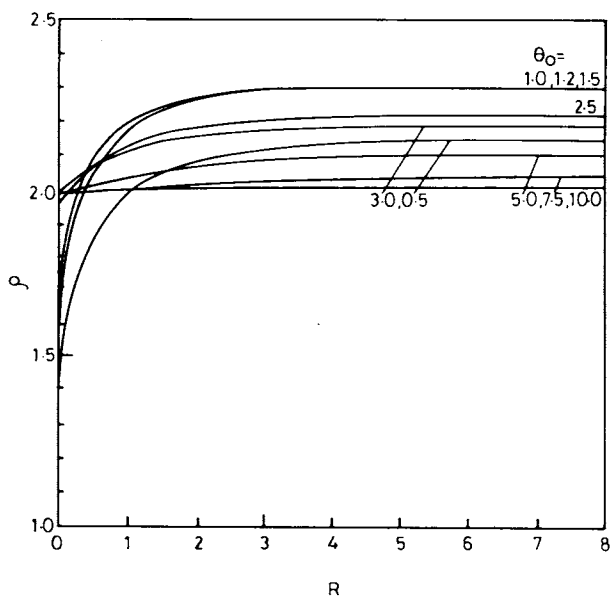


Fig. 13. ρ at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 265^\circ\text{C}$.

When compared to Figures 2–4, it is observed that the use of as low a mono-functional acid concentration as 1% leads to a significant lowering of the number-average chain length and an increase in the monomer conversion, even without a recycle stream. However, the increase in the monomer conversion

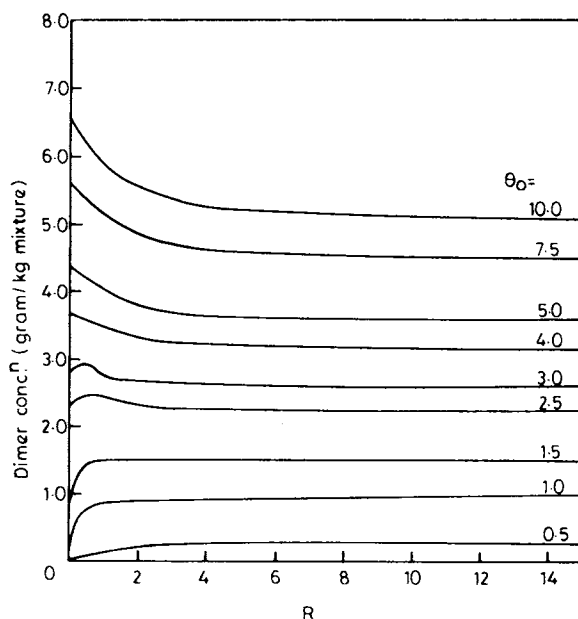


Fig. 14. Cyclic dimer concentration at point 3 for different R and θ_0 . $[W]_1 = 0.44$ mol/kg, $[C_1]_1 = 8.8$ mol/kg, $[A_1]_1 = 0$, $T = 265^\circ\text{C}$.

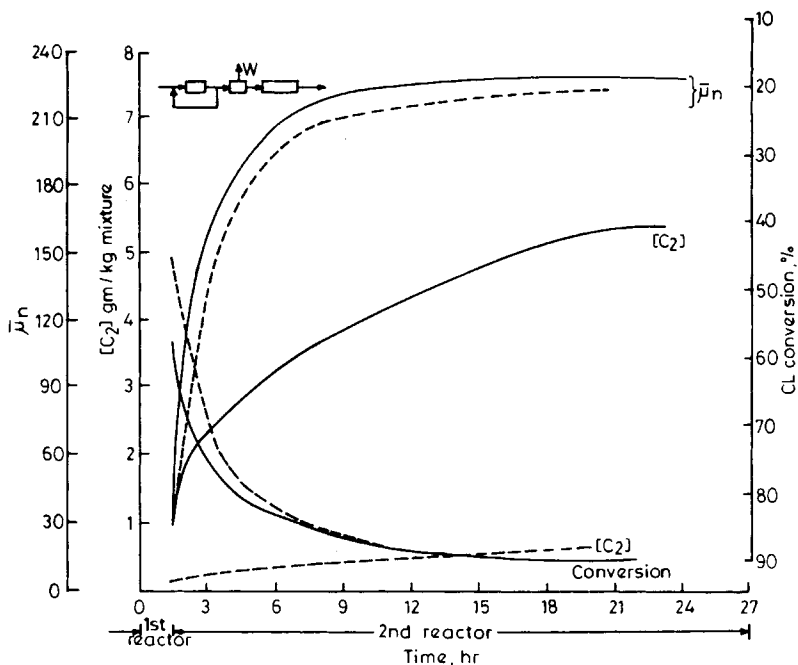


Fig. 15. Conversion of monomer, $\bar{\mu}_n$, and $[C_2]$ as a function of the position in the second reactor of Figure 16 with $R = 0.5$ (—) and $R = 0$ (---). $T = 265^\circ\text{C}$, $[W]_1 = 0.44$ mol/kg, $[A_1]_1 = 0$, $[C_1]_1 = 8.8$ mol/kg, θ_0 in first reactor = 1.5 h.

due to the use of a recycle is observed to be very minimal for the feed conditions studied. This is because the reaction between monofunctional molecules and S_n leads to the additional production of water, as seen from Table I, which speeds up the ring opening reaction. The high values of the polydispersity index of the overall polymer formed in the presence of monofunctional acids is similar to earlier^{4,5,22} results in PFRs as well as in HCSTRs and is apparently because the monofunctional molecules have a different distribution than the bifunctional molecules.

Figures 11–13 show similar results at a higher temperature of 265°C . Higher $\bar{\mu}_n$ and monomer conversions are obtained for the values of θ_0 considered. Once again, for certain values of θ_0 , a recycle reactor is found to give higher monomer conversions than both a PFR as well as an HCSTR. Figure 14 shows values of the cyclic dimer concentration for the same conditions. It is observed that, for $\theta_0 = 1.5$ h, the use of a recycle does *not* lead to a simultaneous increase in the concentration of this undesirable side product which causes problems in the spinning process, and which must be removed from the product by an energy-intensive hot-water extraction. It is thus observed that the use of a recycle stream under certain conditions offers several advantages in nylon 6 polymerization.

Figure 15 shows the variation of monomer conversion, $\bar{\mu}_n$, and $[C_2]$ as a function of time for the reactor configuration shown in Figure 16. For this, $[C_1]_1 = 8.8$ mol/kg, $[W]_1 = 0.44$ mol/kg, $[A_1]_1 = 0$, $R = 0.5$ and 0 , $\theta_1 = 1.5$ h, $T_1 = 265^\circ\text{C}$ (corresponding to the peak in Fig. 11) and the product from this recycle reactor,

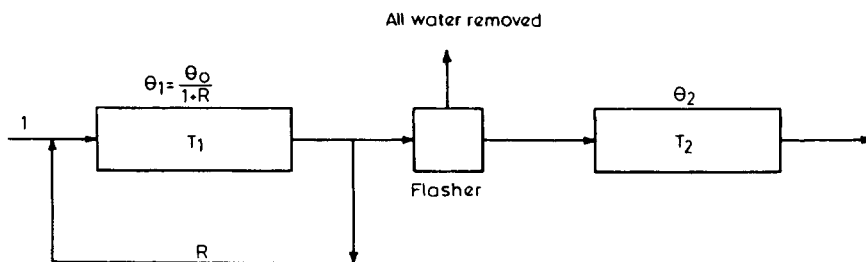


Fig. 16. Another reactor configuration.

after going through a flashing unit where *all* the water present in the reaction mass is removed instantaneously, goes through another isothermal PFR where no water is removed. It is observed that the *final* monomer conversion is almost the same, independent of the recycle; however, slightly higher final values of $\bar{\mu}_n$ are obtained for the case when the recycle stream is present. This increased $\bar{\mu}_n$, however, is accompanied with an increase in the product $[C_2]$. Thus, even though there are several advantages offered by the recycle reactor alone, its combination with a flash unit and a finishing reactor as shown in Figure 16 does not appear as attractive. A detailed study of other similar schemes is called for to obtain optimal combinations of various design variables like recycle ratio, θ_0 , θ_2 , etc.

CONCLUSIONS

It has been shown that, under certain conditions, the use of a recycle stream with a PFR leads to a higher monomer conversion and lower amounts of cyclic compounds without affecting the average chain-length of the polymer formed. In addition, the ideal PFR + recycle model can be used to account for the effects of residence-time distribution in *real* tubular reactors with complex internals. This study also suggests the possibility of the optimization of nylon 6 reactors, with recycle as well as instantaneous flashing of water.

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