Advances in Optimal Control of Polymerization Reactors

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Optimal control of polymerization reactors is of immense interest due to necessity of operating these reactors optimally to produce a polymer product with desired quality characteristics. In the past, various classical methods have been reported for optimal control of polymerization reactors. However, stochastic and evolutionary optimization methods are found effective in solving complex optimization problems due to their flexibility, ease of operation and global perspective. This chapter deals with the recent advances in optimal control of polymerization reactors. Different multistage dynamic optimization strategies based on iterative dynamic programming (IDP), differential evolution (DE) and tabu search (TS) that seek global optimum of more difficult engineering problems are presented. The optimal control performance of these strategies are evaluated and analyzed through their application to a semi-batch styrene-acrylonitrile (SAN) copolymerization reactor.

Keywords: Pontryagin’s maximum principle; iterative dynamic programming; differential evolution, tabu search

1. Introduction

Ensuring product quality is a major issue in a polymerization process since the molecular or morphological properties of a polymer product strongly affect its physical, chemical, thermal, rheological and mechanical properties as well as polymer applications. Polymers of varying grades are usually produced in batch and semi-batch reactors. The properties of the polymers produced in these reactors are closely related to the operating conditions specified for the reactors. The quality of polymer product is assessed by its processability, strength and stiffness. Variations in monomer compositions can result in products with poor physical and mechanical properties. Batch to batch process variations can result in off-specification products. Variations in copolymer composition influence the end use properties of the final product with respect to its flexibility, strength and glass transition temperature. Significant deviation in molecular weight (MW) and molecular weight distribution (MWD) affects the important end use properties such as viscosity, elasticity, strength, toughness and solvent resistance.

The determination of open-loop time varying control policies that maximize or minimize a given performance index is referred to as optimal control/dynamic optimization. These optimal control policies that ensure the satisfaction of the product property requirements and the operational constraints can be calculated off-line, and are implemented on-line such that the system is operated in accordance with these control policies. There has been tremendous interest on the optimal control/dynamic optimization of polymerization reactors and in the past various techniques have been reported for this purpose. [1-6] Most of the studies on optimal control of polymerization reactors are based on classical methods of solution such as Pontryagin’s maximum principle[7] and this method has been widely used to solve the optimal control problems of polymerization reactors.

When the process model is complex and its dimension is large, it becomes tedious to compute the optimal control policies by conventional optimal control techniques. Classical methods like Pontryagin’s maximum principle requires good initial guess for the control inputs and the rate of convergence of the solution is very sensitive to this guess. In case of complex systems, the search space quite often becomes very narrow and choosing the initial guess for the search region requires tedious effort. To overcome these problems, different other techniques have been proposed for optimal control of polymerization reactors. A two step method has been used to compute the reactor temperature and time profiles in a free radical polymerization reactor in order to obtain a polymer with a prescribed molecular weight distribution.[8] This method formulates the control problem into a parameter searching problem that involves tedious numerical computations to solve the nonlinear algebraic equations. The other classical technique that has been used to solve optimal control problems is the control vector parameterization technique. This technique has been used to find the optimal jacket temperature profile for maximizing the productivity in a batch emulsion copolymerization reactor.[9] The control vector parameterization technique requires a set of trial functions with weights to pre-specify the functional form of the control and a deep physical insight into the process behavior.

Dynamic programming developed by Richard Bellman[10] is another powerful method that has been used to solve optimal control problems.[11] This method has the attractive feature of breaking up a complex optimization problem into a number of simpler problems. The stage-by stage calculations involved in this method leads to find the global optimum of the process. However, the difficulty with this method is the problem of setting up the grid values for state and control and when this method is used for optimal control, the dimension of the problem expands drastically limiting its application to the problems of very low dimension. To avoid this difficulty, Luss[12] proposed a technique known as iterative dynamic programming (IDP) that uses accessible state grids and region contraction to reduce dimension expansion. IDP is a popular derivative free method that has been applied to solve different optimal control problems.
including polymerization reactors.\textsuperscript{[17,18]} This method attempts to generate a number of random control policies about the current best rather than maintaining a high dimensional lattice. Though this method is reliable in finding the global optimum and can be applied to problems where the objective function is non-differentiable, it has low efficiency and computationally time consuming.

Optimal control provides considerable scope to develop more efficient and reliable methods for dynamic optimization of polymerization reactors. Of recent, various stochastic and evolutionary optimization algorithms such as genetic algorithm (GA), simulated annealing (SA), differential evolution (DE), ant colony optimization (ACO), tabu search (TS) and artificial neural network (ANN) are widely used to solve optimization problems of different engineering systems.\textsuperscript{[19-27]} These algorithms have an advantage over conventional gradient-based search procedures because they are capable of finding global optima of multi-modal functions and searching design spaces with disjoint feasible regions. This chapter deals with the recent developments in optimal control of polymerization reactors. Main focus is on stochastic and evolutionary optimization strategies that are found effective in solving complex optimization problems due to their flexibility ease of operation, and global perspective. These include dynamic process model assisted multistage dynamic optimization strategies based on iterative dynamic programming (IDP), differential evolution (DE) and tabu search (TS). The performance of these strategies for optimal control of polymerization reactors is evaluated and analyzed with their applications to a semi-batch styrene-acrylonitrile (SAN) copolymerization reactor.

2. Optimal control problem

The major objective in optimal control is to determine the time varying open-loop control policies that maximize or minimize the objective function which specify the process performance as a function of process variables and their changes. The process performance can stated to be either achieving a desired product quality or maximizing the product yield of a batch process. The general open-loop optimal control problem with fixed terminal time, considering a lumped parameter batch/semi-batch process can be stated as follows.

Find a control vector $u(t)$ over $t_f[t_0, t_f]$ to maximize (minimize) a performance index $J(x,u)$:

$$J(x,u) = \int_0^{t_f} \phi(x(t), u(t), t) dt$$

Subject to

1. $\dot{x}(t) = f(x(t), u(t), t)$, $x(t_0) = x_0$ (2)
2. $h(x(t), u(t), t) = 0$ (3)
3. $g(x(t), u(t), t) \leq 0$ (4)
4. $x^L \leq x(t) \leq x^U$ (5)
5. $u^L \leq u(t) \leq u^U$ (6)

where $J$ is the performance index, $x$ is the vector of state variables, $u$ is the vector of control variables. Eq. (2) is the system of ordinary differential equations with their initial conditions, Eqs. (3) and (4) are the equality and inequality algebraic constraints and Eqs. (5) and (6) are upper and lower bounds in the state and control variables.

3. Multistage dynamic optimization

A multistage problem arises due to the natural extension of a single stage optimization to a system of more stages in which the output from one stage is input to the subsequent stage. The procedure for multistage optimization can be referred elsewhere.\textsuperscript{[28]} Multistage optimization problems require special techniques to break them into computationally manageable units. Various methods such as the dynamic programming based on principle of optimality of Bellman\textsuperscript{[14]} and the methods derived from calculus of variations and Pontryagin’s Maximum principle\textsuperscript{[27]} have been developed for optimizing the performance of dynamic systems. Multistage dynamic optimization is an approach used for stage wise optimization of complex dynamic systems. In this approach, the optimal control problem is considered by dividing the entire batch duration into finite number of time instants referred to as discrete stages. The control variables and the corresponding state variables that satisfy the objective function are evaluated in stage wise manner.

Discretize the process into $N$ stages. Define $f^i$, $u^i$ and $x^i$ as the objective function, control vector and state vector, respectively, for stage $i$. Here, the procedure adopted for solving the optimal control problem is similar to that of the dynamic programming based on the principle of optimality.\textsuperscript{[14]} This procedure is briefly in the following steps:

1. The optimum value of the objective function, $f^i[x^i]$ for stage 1 driven by the best control vector $u^i$ along with the state vector $x^i$ is represented as

$$f^i[x^i] = \min_{u^i} f^i[x^i, u^i]$$

where $J$ is the performance index, $x$ is the vector of state variables, $u$ is the vector of control variables. Eq. (2) is the system of ordinary differential equations with their initial conditions, Eqs. (3) and (4) are the equality and inequality algebraic constraints and Eqs. (5) and (6) are upper and lower bounds in the state and control variables.
2. The value of the objective function, \( f^2[x^2] \) for stage 2 is determined based on the best control vector \( u^2 \) along with the state vector \( x^2 \) as given by

\[
f^2[x^2] = f^1[x^1] + \min_{u^2} f^2_o[x^2, u^2]
\]  

(8)

3. Recursive generalization of the above procedure for the \( k^{th} \) stage is represented by

\[
f^k[x^k] = f^{k-1}[x^{k-1}] + \min_{u^k} f^k_o[x^k, u^k]
\]  

(9)

In this procedure, \( f^k_o \) represents the performance index of stage \( k \).

4. Optimal control algorithms considered

The algorithms that are used to derive multistage dynamic optimization strategies for polymerization reactors are described.

4.1 Iterative dynamic programming

Iterative Dynamic Programming (IDP) is one of the leading direct search techniques for solving optimal control problems.\(^{[16]}\) IDP attempts to overcome the curse of dimensionality associated with the dynamic programming by generating a number of random control policies about the current best, rather than maintain a complete high dimensional lattice. In this work, IDP based strategy is used for multistage dynamic optimization of polymerization reactor. The optimal control problem is to find the control \( u(t) \) in the time interval \( t_{k-1} \leq t \leq t_k \), so as to satisfy the desired objectives. The optimal control policy is approximated by a piecewise constant control policy over \( P \) time stages, each of length \( L = t_f/P \), so that the time interval \( t_{k-1} \leq t \leq t_k \) is considered with the constant control \( u(t) = u(k-1) \). The problem then is to find \( u(0), u(1), \ldots, u(P-1) \) that satisfy the performance index. More details on IDP algorithm and its implementation procedure can be referred elsewhere.\(^{[16]}\)

4.2 Differential evolution (DE)

Among the evolutionary algorithms, differential evolution (DE) is a simple population based search algorithm for global optimization of real valued functions.\(^{[21]}\) Its robustness and effectiveness has been demonstrated in a variety of applications.\(^{[22]}\) DE is similar to Genetic Algorithm (GA) in the sense that it uses the same evolutionary operators like selection, recombination and mutation as in GA, however, the significant difference is that DE uses distance and direction information from the current population to guide the search process. DE is controlled by three parameters, namely, population size (NP), crossover operator (CR) and mutation constant called scaling factor (F). The flow chart of DE algorithm is shown in Fig. 1. The detailed description of DE is found elsewhere.\(^{[22]}\)

![Flow chart of differential evolution algorithm.](image)
4.3 Tabu search algorithm

Tabu search (TS) is a meta-heuristic problem solving approach that incorporates adaptive memory and responsive exploration, which makes it to penetrate complexities that often confound alternative approaches. The adaptive memory feature of TS allows the implementation of procedures that are capable of searching the solution space economically and effectively. Responsive exploration integrates intelligent search mechanisms to exploit good solution features while exploring new promising regions.

TS begins by choosing an initial solution $x_0$. The neighbor solutions are generated by modifying the existing solution through a sequence of moves. The best new neighbor, $x^*$ is used as the starting point for the next iteration unless it is in Tabu list. Thus, even if no neighbor solutions are better than the initial solution, the best solution is still chosen as the starting point for the next iteration. A record of the best solutions ever found, $x^*$ is separately maintained. In addition, the adaptive memory in tabu list guides the search process by taking the advantage of historical information. This memory enables TS to make strategic choices and achieve responsive exploration. Tabu search algorithm involves various computational elements which include neighbors generation and neighborhood search, tabu list, short term memory and long term memory, intensification and diversification, aspiration criterion and stopping criteria. The flow chart of the TS algorithm is shown in Fig. 2. The detailed description of tabu search can be seen elsewhere.

![Flow chart of the TS algorithm.](image)

5. Application process for optimal control

Styrene acrylonitrile (SAN) copolymer is commercially important and optimal control of the reactor is needed to achieve the desired polymer properties. In this work, the process of solution copolymerization of styrene and acrylonitrile in a semi-batch reactor is considered for implementation of the proposed dynamic optimization strategies. Xylene and AIBN are used as solvent and initiator. The feed is a mixture of monomers, solvent and initiator which enters the reactor in semi-batch mode. Initial volume of the reactor is 1.01 l, and the initial design parameters are the solvent mole fraction $f_x = 0.25$, and initiator concentration $I_0 = 0.05$ mol/l. The mol ratio of monomers in the feed, $M_1/M_2$ is 1.5.

5.1 Mathematical model

The following kinetic model is used to describe the homogeneous solution free-radical copolymerization of styrene with acrylonitrile.
Initiation:
\[ I \longrightarrow 2R \]
\[ R + M_1 \overset{k_{1i}}{\longrightarrow} P_{10} \]  
\[ R + M_2 \overset{k_{1i}}{\longrightarrow} Q_{01} \]  
\( k_{1i} \) is the initiation rate constant.

Propagation:
\[ P_{n0} + M_1 \overset{k_{2i}}{\longrightarrow} P_{n+1,0} \]  
\[ Q_{n0} + M_2 \overset{k_{2i}}{\longrightarrow} Q_{n+1,0} \]  
\( k_{2i} \) is the propagation rate constant.

Combination termination:
\[ P_{n0} + P_{m0} \overset{k_{ter}}{\longrightarrow} M_{n+m,0} \]  
\[ Q_{n0} + Q_{m0} \overset{k_{ter}}{\longrightarrow} M_{n+m,0} \]  
\( k_{ter} \) is the combination termination rate constant.

Disproportionation termination:
\[ P_{n0} + Q_{m0} \overset{k_{ter}}{\longrightarrow} M_{n,m} + M_{n,m} \]  
\( k_{ter} \) is the disproportionation termination rate constant.

Chain Transfer:
\[ P_{n0} + M_1 \overset{k_{t}}{\longrightarrow} P_{n-1,0} + f \]  
\[ Q_{n0} + M_2 \overset{k_{t}}{\longrightarrow} Q_{n-1,0} + f \]  
\( f \) is the chain transfer agent.

where \( P_{n0} \) represents a growing polymer chain with \( n \) units of monomer 1 (styrene) and \( m \) units of monomer 2 (acrylonitrile) with monomer 1 on the chain end. Similarly, \( Q_{n0} \) represents a growing copolymer chain with monomer 2 on the end. The \( M_{n,m} \) denotes inactive or dead polymer.

The copolymer molecular weight (MW) and molecular weight distribution (MWD) are computed using three leading moments of the total number average copolymers. The instantaneous \( k^\text{th} \) moment is given by:
\[ \lambda_k = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} (w_1 + w_2)^k M_{n,m} \]  
where \( w_1 \) and \( w_2 \) are the molecular weights of styrene and acrylonitrile, respectively. The total number average chain length (\( X_n \)), the total weight average chain length (\( X_w \)) and the polydispersity index (PD) are expressed as:
\[ X_n = \lambda_n^d / \lambda_0^d \]  
\[ X_w = \lambda_n^d / \lambda_0^d \]  
\[ PD = X_w / X_n \]  
\( k_{ter} \) is the combination termination rate constant.

The modeling equations of the semi-batch copolymerization reactor are given as follows:
\[ \frac{dM_1}{dt} = u(M_{1i} - M_1) - k_{21}Q_0 + k_{12}Q_0 \]  
\[ \frac{dM_2}{dt} = u(M_{2i} - M_2) - k_{22}Q_0 + k_{12}Q_0 \]  
\[ \frac{dQ}{dt} = u(1 - I) - k_0I \]  
\[ \frac{dV}{dt} = u \]  
The polymer live moments are given by:
\[ P = \left( \frac{V}{k_{21}} \right)^{(k_{21} + k_{12})} + 2\beta(k_{21} + k_{12}) + \beta^2(k_{22} + k_{12}) \right)^{1/2} \]  
where \( \beta = \left( k_{21} + k_{22} \right) / \left( k_{12} + k_{22} \right) \) \( \Phi \) is the polymerization rate constant.

Pseudo state approximation to live polymers leads to the following live polymer moment equations:
\[ P_1 = \left[ w_1C + \alpha Q \right] / \gamma_1 + w_1 \left( \alpha P + \alpha Q \right) / \gamma_1 \right] / \left( 1 - \alpha \right) \]  
\[ Q_1 = \left[ w_1C + \alpha Q \right] / \gamma_2 + w_1 \left( \alpha P + \alpha Q \right) / \gamma_2 \right] / \left( 1 - \alpha \right) \]  
\[ P_2 = \left[ w_1C + \alpha Q \right] / \gamma_3 + w_1 \left( \alpha P + \alpha Q \right) / \gamma_3 \right] / \left( 1 - \alpha \right) \]  
\[ Q_2 = \left[ w_1C + \alpha Q \right] / \gamma_4 + w_1 \left( \alpha P + \alpha Q \right) / \gamma_4 \right] / \left( 1 - \alpha \right) \]  
where \( \gamma_1 = k_{21}C + k_{22}Q / k_{12}M \) ; \( \gamma_2 = k_{22}C + k_{22}Q / k_{12}M \) ; \( \gamma_3 = k_{22}C + k_{22}Q / k_{12}M \) ; \( \gamma_4 = k_{22}C + k_{22}Q / k_{12}M \) ; \( k_{21} \) is the combination termination rate constant.

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The constraints on operating variables are chosen based on the requirements for reaction rate, heat transfer, and conversion of monomer 1 (styrene) is defined as:

$$\alpha_1 = \frac{k_p M_1}{(k_p + k_{11})M_1 + (k_p + k_{12})M_2 + (k_p + k_{111})P + (k_{112} + k_{122})Q}$$

$$\alpha_2 = \frac{k_p M_2}{(k_p + k_{11})M_1 + (k_p + k_{12})M_2 + (k_p + k_{111})P + (k_{112} + k_{122})Q}$$

The moment equations for dead polymers are given by:

$$\frac{dk_1}{dt} = (k_{11} P + k_{111} M + k_{112} M + k_{112} M)\rho + (k_{112} M + k_{122} M)Q - (k_{11} P + k_{111} Q)$$

$$\frac{dk_2}{dt} = (k_{12} P + k_{121} M + k_{122} M + k_{122} M)\rho + (k_{122} M + k_{122} M)Q - (k_{12} P + k_{121} Q)$$

The rate constants, $k_{1j}$, $k_{pj}$, $k_{1pi}$ and $k_{adj}$ are Arrhenius functions of temperature, the pre-exponential factors and activation energies of which are reported elsewhere.\[29\]

The instantaneous copolymer composition $F_i$ is determined by the relative reactivities of the monomers ($r_1$ and $r_2$) and the bulk phase monomer mol fractions ($f_i$ and $f_j$) as given by:

$$F_i = (r_i f_i + f_i f_j) / (r_i f_i + 2 f_i f_j + r_j f_j)$$

The conversion of monomer 1 (styrene) is defined as:

$$x_i = \left[ \frac{[M_{2u} + \int_0^t \dot{M}_{2u}(t) dt - \int_0^t \dot{V}(t) dt]}{[M_{2u} + \int_0^t \dot{V}(t) dt]} \right]$$

This model is in the general form of Eq (1), where the set of state variables, $X$ and the control vector $U$ are given by $X(t) = [M_1(t), M_2(t), I(t), F(t), \lambda_d(t), \lambda_i(t), \lambda_2(t)]^T$ and $U(t) = [I(t), \dot{M}(t)]^T$.

In the above equations, $M_1$ and $M_2$ are the molar concentrations of the unreacted monomers (styrene and acrylonitrile) at time $t$, $I$ is the concentration of the unreacted initiator, $F$ is the volume of the reaction mixture, $\lambda_k (k = 0, 1, . . . )$ is the $k$th moment of molecular weight distribution of the dead copolymer, $T$ is the temperature of the reaction mixture, and $\dot{u}$ is the volumetric flow rate of the feed mixture to the reactor. More details of reaction kinetics and numerical data pertaining to this system can be referred elsewhere.\[29\]

5.2 Control objectives

Optimal control of SAN copolymerization involves the determination of control policies to maintain the specified objectives while satisfying the constraints. Considering the wt% of acrylonitrile in the polymer to be in the range of 25-35%, the desired values of copolymer composition ($F_i(t)$) and number average molecular weight ($MWD$) are chosen to be 0.58 and 30,000, respectively. The objectives are specified as minimizing the deviations of copolymer composition, $F_1$ and molecular weight, $MWD$ from their respective desired values during the entire span of the reaction. For satisfying these objectives, monomer addition rate, $u$ and reactor temperature, $T$ are selected as control variables. If only one polymer quality parameter is controlled by manipulating one control variable, uncontrolled property parameters may deviate from their desired values as the reaction proceeds. Therefore simultaneous optimization of desired polymer quality characteristics is desirable by manipulating the decision variables.

The optimal control problem is considered as optimizing the single objectives as well as both the objectives simultaneously. The objectives of SAN copolymerization are formulated as

$$J_1 = [1 - MWD(t)/MWD]^2$$

$$J_2 = [1 - F_1(t)/F_1(D)]^2$$

$$J_3 = [(1 - F_1(t)/F_1(D))^2 + [1 - MWD(t)/MWD]^2$$

Here $MWD$ and $F_1$ are the molecular weight and copolymer composition, and $MWD$ and $F_1$ are their respective desired values. The notation $t$ here refers discrete time. The hard constraints are set as

$$0 \leq u(t) \leq 0.07 \text{ (l/min)}$$

$$320 \leq T(t) \leq 368 \text{ (k)}$$

$$\dot{V}(t) \leq 4.01$$

These constraints on operating variables are chosen based on the requirements for reaction rate, heat transfer limitation and reactor safety.

5.3 Discrete control sequences

The optimal control problem of SAN copolymerization reactor is considered as a problem of dynamic optimization by dividing the entire span of reaction time into finite number of time instants which can also be referred as discrete stages. The total duration of reaction is fixed at 300 min. Thus $u$ and $T$ profiles within the ranges of their constraints for the
entire duration of reaction are equally discretized into 19 stages having 20 time points. These discrete control sequences for feed flow and temperature are given by
\[ u = [u_1, u_2, \ldots, u_{20}]^T \]
\[ T = [T_1, T_2, \ldots, T_{20}]^T \]

In single objective optimization problems, the objective is to determine the temperature \( T \) policy to maximize \( MW \) defined by the objective function in Eq. (23) and monomer feed rate \( (u) \) policy to maximize \( F_t \) defined by the objective function in Eq. (24). In simultaneous optimization problem, the objective is to determine both the \( T \) and \( u \) policies to satisfy the objective function defined in Eq. (25).

6. Optimal control of styrene acrylonitrile (SAN) copolymerization reactor using iterative dynamic programming (IDP) and differential evolution (DE)

The optimal control problem is to find the control input \( u(t) \) in the time interval \( t_k; 1 \leq t \leq t_k \), so as to satisfy the desired objectives. The optimal control policy is approximated by a piecewise constant control policy over \( P \) time stages, each of length \( L \), where \( L = t_f / P \), so that the time interval \( t_k; 1 \leq t \leq t_k \) is considered with the constant control \( u(t) = u(k-1) \). The problem then is to find \( u(0), u(1), \ldots u(P-1) \) that satisfy the performance index.

6.1 Implementation procedure for IDP

A multistage dynamic optimization strategy based on iterative dynamic programming (IDP) is employed to determine the optimal control policies for temperature \( T \), feed rate \( u \) and for both \( T \) and \( u \). The algorithm of iterative dynamic programming (IDP) consists of the following steps.[30]

1. Divide the time interval \( t_f \) into \( P \) stages, each of length \( L \).
2. Choose the number of \( x \)-grid points \( N \), the number of allowable values \( M \) for the control \( u \), the region \( r \) for the control values, the region contraction factor \( \gamma \) and the region restoration factor \( \eta \).
3. Choose the number of iterations used in every pass and the number of passes. Set the pass number index \( q = 1 \) and the iteration number index \( j = 1 \).
4. Set \( r^j = r_0 \) where \( r_0 \) is initial region size.
5. By choosing \( N \) values of control inside allowable region, integrate Eq. 3 representing the polymerization reactor model in Section 5.1 for \( N \) times to generate the \( x \)-grid at each time stage.
6. Starting at the last time stage \( P \), corresponding to time \( t_f - L \), for each \( x \)-grid point, integrate the model equations from \( t_f - L \) to \( t_f \) for all the \( M \) allowable values of control \( u \) or \( T \). Choose the control that satisfies the performance index and store the value of control as \( u^k \) for use in the next step of the algorithm.
7. Step back to stage \( P-1 \), corresponding to time \( t_f - 2L \), and integrate the model equations from \( t_f - 2L \) to \( t_f \) for each \( x \)-grid point with the \( M \) allowable values of control. To continue integration from \( t_f - L \) to \( t_f \), choose the control from step 6 that corresponds to the grid point closest to the resulting \( x \). Compare the \( M \) values of the performance index and store the values of control, \( u^k \) that gives the desired optimum value.
8. Continue the procedure until stage 1, corresponding to the initial time \( t=0 \) is reached. Store the control policy that satisfies the performance index and store the corresponding \( x \)-trajectory.
9. Reduce the search region for allowable control values by a factor \( \gamma \):
\[ r^{j+1} = \gamma r^j \]
(30)
The updated control trajectory is
\[ u_{j+1}^i(P) = u_j^i(P) + Dr^j \]
(31)
where \( u_j^i(P) \) is the optimum value obtained in the previous iteration and \( D \) is a diagonal matrix of random numbers varying from \(-1 \) to \( 1 \).
10. Increment the iteration index \( j \) by 1 and go to step 3. Continue the procedure for a specified number of iterations for each pass.
11. Increment the pass number index \( q \) by 1 and go to step 4. Continue the procedure for a specified number passes.

6.2 Implementation procedure for DE

DE is implemented to compute the optimal control policies using the following procedure.[28]

1. Divide the time interval \( t_f \) into \( P \) stages, each of length \( L \). Set the DE parameters \( D, NP, CR \) and \( gen_{max} \), where \( D \) refers the number of control inputs and \( gen_{max} \) refers the maximum generations.
2. Initialize the vectors of the population representing the control input randomly for all stages:
\[ X_{i,j} = \text{rand}(0,1),(b_{ij,1} - b_{ij,2}) + b_{ij,2} , i=1,\ldots, NP \ ; j=1,\ldots, D \]
(32)
3. Set the stage index \( P=1 \) and stage time \( t=t_1 \).
4. Set the generation index \( q=1 \).
5. Integrate the process model, Eq. (3) representing the polymerization reactor model in Section 5.1 from the initial time to the end time of the current stage for each of the NP population of the control input (u or T) and evaluate the objective function values at the end of the stage.

6. Perform mutation, crossover and selection based on the objective function values of trial and target vectors.

7. Set q=q+1. Go to step 5 and continue the procedure.

8. Determine the optimal control input, \( u^*(P) \) based on the convergence in objective function or satisfying the specified number of generations. Store the converged values of the control input, state vector and objective function at the end of current stage for use in the next stage calculation.

9. Set the stage number \( P=P+1 \) and stage time \( t=t_2 \). Go to step 4 and continue the procedure. The evaluation of objective function values for this stage is based on the model integration using the optimal control input for the first stage duration along with the integration for NP control input population of the second stage using the optimal state of the first stage end as the starting point.

10. Go to step 4 and repeat the procedure for all stages. The resulting control input of each stage represents the optimal trajectory.

Implementation scheme of DE for multistage dynamic optimization of copolymerization reactor is shown in Fig. 3.

6.3 Analysis of results

Optimal control of SAN copolymerization reactor is carried out by using DE and IDP strategies. The IDP strategy is used to computes the optimal control policies iteratively and recursively from the last stage to first stage using the procedure described in section 6.1. The control values, \( M \) for each grid point, the region restoration factor, \( \eta \) and the region contraction factor, \( \gamma \) are selected appropriately. The diagonal matrix, \( D \) is generated randomly within the range of -1 to 1. The initial temperature policy for all stages is set as 3330K. The parameters for temperature policy are set as \( P=100, \ r_m=15, \ \gamma=0.85 \) and \( \eta=0.1 \). The initial feed rate policy for all stages is set as \( P=50, \ r_m=5, \ \gamma=0.75 \) and \( \eta=0.5 \). The parameters for simultaneous optimization are set as \( P=100, \ r_m=15, \ \gamma=0.85 \) and \( \eta=0.1 \). The model integration for each stage is performed with a time step of 1 min using the

![Fig. 3. Implementation scheme of DE for dynamic optimization.](image-url)
procedure given in section 6.1. The optimal solution for single control policies as well as dual control policies is achieved with in 5 passes, each pass with 50 iterations.

For DE implementation, the control input at the beginning of the first stage is chosen at the lower bound of the input space. The process model is integrated for the duration of 15 min with a time step of 1 min from the beginning to the end of the first stage and the objective function values are evaluated for all the control input population at the end of this stage. The optimal control policies for temperature \( T \); feed rate \( u \) and for both \( T \) and \( u \) are determined by setting the parameters of DE as \( NP=100, \ CR=0.6, \ F=0.7 \) for \( T \) policy, \( NP=20, \ CR=0.6, \ F=0.4 \) for \( u \) policy, and \( NP=400, \ CR=0.6 \) and \( F=0.5 \) for \( T \) and \( u \) policies. The DE operations of mutation, crossover and selection are implemented iteratively to minimize the objective function and to determine the best control input for this stage. For second stage solution, random population is generated for the control input at the end of the second stage. The model is integrated from the beginning to the end of the first stage based on the optimal control input of the first stage and from the beginning to the end of the second stage based on the control input population of the second stage. Iterative convergence of the objective function leads to determine the optimal control input for the second stage. This procedure is continued until the end of last stage. The control input values thus determined at the end of each stage represents the optimal control policy.

The single control policies for \( T \) and \( u \) are determined while satisfying the objective functions specified in Eqs. (23) and (24), respectively. For simultaneous optimization, DE implementation at each stage considers the combination of the control input populations of \( T \) and \( u \). The optimal solution for single control policies as well as dual control policies is achieved with in 50 iterations. For the case of single objective optimization of keeping \( MW \) at its desired value based on optimal \( T \) policy, the DE strategy is found to provide effective control with improved polydispersity. The \( MW \) obtained by IDP is found to deviate considerably from the desired value. The uncontrolled copolymer composition (\( F_1 \)) is found to deviate from its desired value when only \( MW \) is controlled. When copolymer composition (\( F_1 \)) is controlled at its desired value with optimal \( u \) policy, the uncontrolled \( MW \) is found to deviate considerably from its desired value. The copolymer composition control results show that the DE and IDP display similar trend in response behaviors. The dual control policies of \( T \) and \( u \) determined by IDP and DE are depicted in Figs 4. The process responses of polydispersity and conversion corresponding to the dual control policies of these methods are shown in Fig. 5. These results show that both the temperature and feed rate policies of DE maintain \( MW \) and \( F_1 \) almost near to their desired values. However, the results of IDP exhibit deviation than those of DE. The \( MW \) values obtained at the end of the reaction by DE and IDP are 29372 and 28981, respectively, and the corresponding \( F_1 \) values obtained at the end of the reaction are 0.5712 and 0.5564, respectively. These results show that the DE policies better achieve the desired objectives over IDP policies. DE has the feature that it extracts distance and direction information from the current control input vectors and adds random deviation for diversity to generate new parameter vectors through the operation of mutation, crossover and selection. These features make DE to be more effective in determining the optimal control policies to achieve the desired objectives.

### 6.3.1 Computational efficiencies

The computational efficiencies and the normalized absolute error values of DE and IDP are evaluated. These results show that the execution times required for the iterative convergence of the optimal control policies by DE is much lower to that of IDP. The performance of these methods is also evaluated based on the normalized absolute error which is defined as the difference between the values of the desired and end objectives. The normalized absolute error values of DE are found to be much lower to that of IDP. The results thus evaluated for single policies and dual policies show the better performance of DE over IDP.

**Table 1. Comparison of the methods of dynamic optimization**

<table>
<thead>
<tr>
<th>Strategy</th>
<th>Control Policy</th>
<th>Computational efficiency</th>
<th>Normalized absolute error</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Convergence time (sec)</td>
<td>Implementation time (sec)</td>
</tr>
<tr>
<td>DE</td>
<td>T</td>
<td>4.68</td>
<td>22.92</td>
</tr>
<tr>
<td></td>
<td>U</td>
<td>6.39</td>
<td>35.62</td>
</tr>
<tr>
<td></td>
<td>T &amp; u</td>
<td>10.72</td>
<td>68.13</td>
</tr>
<tr>
<td>IDP</td>
<td>T</td>
<td>38.59</td>
<td>226.09</td>
</tr>
<tr>
<td></td>
<td>U</td>
<td>26.87</td>
<td>165.00</td>
</tr>
<tr>
<td></td>
<td>T &amp; u</td>
<td>73.90</td>
<td>179.06</td>
</tr>
</tbody>
</table>
7. Optimal control of SAN copolymerization reactor using IDP and Tabu search

Multistage dynamic optimization of polymerization reactor is carried out by using TS\(^{(31)}\) and compared with the IDP strategy. Design and implementation of IDP is same as in section 6.1.

7.1 Implementation of TS

The control objectives and discrete control sequences are briefed in sections 5.2 and 5.3. TS implementation is carried to compute the optimal control policies in polymerization reactor using the procedure described in Section 4.3 and the flow chart of TS algorithm is given in Fig. 2. Initially, the control vector is specified to be a constant value at each of
the 20 points representing 19 stages within the constraints specified by Eq. (26). Since the process operation is considered for a time duration of 300 min, the duration of each stage corresponds to 15 min. TS explores the search space of feasible solutions by a sequence of moves. The control input at the beginning of the first stage is chosen at the lower bound of the input space. For MW optimization, ten neighbors are generated at the end of the first stage by introducing random changes in the search space of T with an incremental variation of -0.4 to 0.4.

The elements of tabu search for computing the optimal control policies in SAN polymerization reactor is set as follows. The sizes of both recency and frequency based tabu lists are fixed as 50. The sizes of these lists are chosen such that they forbid revisiting of unpromising solutions in the search process. An intensification strategy with the format of a sine function is employed. A parameter (b) value of 4.0001 that controls the oscillation period of a sine function is employed. An aspiration criterion based on a sigmoid function with the parameters as $k_{center}=0.3$ and $\sigma=7/M$ with M as specified number of iterations is employed. The number of neighbors for each control input of each stage is set as 10. The process model is integrated for a duration of 15min with a time step of 1 min from the beginning to the end of the first stage and the objective function values are evaluated for all the generated neighbors at the end of this stage. The iterative convergence of TS establishes the best control input ($T$) along with its objective function. This becomes the optimal control point for the first stage, which is then used as a starting point for the second stage solution. For second stage solution, random neighbors are generated at the end of the second stage around the optimal $T$ of the first stage. The model is integrated from the beginning to the end of the first stage based on the initial control point ($T$) and from the beginning to the end of the second stage for each of the neighbors generated at the end of second stage. The optimal control inputs for successive stages until the end of last stage are established in a similar manner. The control input values thus determined at the end of each stage represents the optimal control policy for $T$. For $F_1$ optimization, ten neighbors are generated at the end of each stage with an incremental variation of $-1.0 \times 10^{-6}$ to $1.0 \times 10^{-6}$. In analogous manner, optimal control policy for $u$ is determined adapting the similar TS procedure as in $T$ policy. For establishing dual control policies for $T$ and $u$, multistage dynamic optimization by TS is carried out by considering incremental variations in neighbors generation of $T$ and $u$ within the limits of -0.4 to 0.4, and $-1.0 \times 10^{-6}$ to $1.0 \times 10^{-6}$, respectively. This case requires the evaluation of the objective function values for 100 neighbor combinations at each of the control point representing $T$ and $u$. The implementation of TS strategy for optimal control of SAN copolymerization reactor is shown in Fig. 6.

### Fig. 6. Implementation scheme of TS for dynamic optimization.

#### 7.2 Analysis of results

Tabu search (TS) is designed and applied to determine the optimal control policies that satisfy the individual and multiple objectives of SAN copolymerization reactor. More details on tabu search implantation can be referred elsewhere. The results are further compared with the dynamic optimization results of IDP. For single objective
optimization of maintaining the desired MW with optimal T policy, the method of TS is found to provide better control with improved polydispersity. The MW obtained by IDP is found to differ considerably from the desired value. The uncontrolled copolymer composition \( F_1 \) is found to deviate from its desired value when only MW is controlled. When copolymer composition \( F_1 \) is controlled at its desired value with optimal u policy, the uncontrolled MW is found to deviate considerably from its desired value. The copolymer composition control results show that the TS and IDP exhibits almost similar trend in response behaviors.

Dual control policies of \( T \) and \( u \) for multistage dynamic optimization of polymerization reactor are determined using TS and the results are further compared with the dynamic optimization results of IDP. Fig. 7 depicts the results of the dual control policies of these methods along with the objective function values. The process responses of polydispersity and conversion corresponding to the dual control policies of these methods are shown in Fig. 8. These results show that both the temperature and feed rate policies of TS maintain MW and \( F_1 \) almost near to their desired values. However, the results of IDP are observed to deviate than those of TS. The MW values obtained at the end of the reaction by TS and IDP are 29986, and 28981, respectively, and the corresponding \( F_1 \) values obtained at the end of the reaction are 0.5759, and 0.5564, respectively. These results show the better performance of TS over IDP.

### 7.2.1 Effect of design parameters and computational efficiencies

The performance of TS is further studied with respect to the effect of the parameters \( \sigma \) and \( k_{\text{center}} \) involved in the sigmoid function. The effect of \( k_{\text{center}} \) and \( \sigma \) within their specified ranges is found marginal on the results, whereas the execution time is found to increase with the increase of neighbors. The computational efficiencies and the normalized absolute error values of TS and IDP are also studied. These results show that the execution times required for the iterative convergence of the optimal control policies by TS are much lower to that of IDP. The performance of these methods is also evaluated based on the normalized absolute error which is defined as the difference between the values of the desired and end objectives. These results show the improved performance of TS over IDP. The results of TS and IDP are also evaluated based on computational cost and memory storage. The memory storage requirement of Tabu search and IDP is given in Table 1 along with the computational efficiencies of these methods. The results thus show the better performance of tabu search over IDP. The TS based optimal control strategy is also compared with the differential evolution (DE) based strategy [28]. These results show that TS also has significant advantage over DE in solution quality.

### Table 1. Comparison of the methods of dynamic optimization

<table>
<thead>
<tr>
<th>Strategy</th>
<th>Control Policy</th>
<th>Computational efficiency</th>
<th>Normalized absolute error</th>
<th>Memory storage, k</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Convergence time (sec)</td>
<td>Implementation time (sec)</td>
<td></td>
</tr>
<tr>
<td>TS</td>
<td>T</td>
<td>1.89</td>
<td>16.83</td>
<td>0.000566</td>
</tr>
<tr>
<td></td>
<td>U</td>
<td>2.19</td>
<td>13.81</td>
<td>0.01379</td>
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<td>T &amp; u</td>
<td>4.25</td>
<td>19.21</td>
<td>0.007535</td>
</tr>
<tr>
<td>IDP</td>
<td>T</td>
<td>38.59</td>
<td>226.09</td>
<td>0.3453</td>
</tr>
<tr>
<td></td>
<td>U</td>
<td>26.87</td>
<td>165.00</td>
<td>0.03448</td>
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<tr>
<td></td>
<td>T &amp; u</td>
<td>73.90</td>
<td>179.06</td>
<td>0.0764</td>
</tr>
</tbody>
</table>
8. Conclusions

In this chapter different multistage dynamic optimization strategies based on iterative dynamic programming (IDP), differential evolution (DE) and tabu search (TS) are presented for optimal control of polymerization reactors. The results of these strategies are evaluated by applying them for optimal control of a semi-batch styrene-acrylonitrile (SAN) copolymerization reactor. The comparison of the results of IDP and DE based strategies have shown the better performance of DE based strategy in establishing single and dual control policies to maintain the desired polymer
product quality requirements. Further the comparison of multistage dynamic optimization results of TS and IDP for SAN copolymerization reactor have shown the efficacy of TS for determining the single and dual control policies that satisfy the desired polymer product characteristics. The dynamic process model assisted multistage dynamic optimization strategies based on TS and DE are found effective for solving complex optimization problems in polymerization reactors.

Notation

- $I_i$: initiator concentration in feed
- $k_d$: initiator decomposition rate constant
- $k_{ij}$: chain transfer rate constant of species $i$, $j$
- $k_p$: propagation rate constant species $i$, $j$
- $k_{cij}$: combination termination rate constant of species $i$, $j$
- $k_{dij}$: disproportionation termination rate constant of species $i$, $j$
- $M_i$: monomer concentration in reaction in feed of species $i$
- $P$: Total growing polymer concentration of type-1 [mol/l]
- $P_i$: moment of total number $MWD$ of radicals of type-1
- $Q$: total growing polymer concentration of type
- $Q_i$: moment of total number $MWD$ of radicals of species $i$
- $r_{ij}$: copolymerization reaction rate
- $t_i$: manipulated variable of species $i$
- $V$: reactor volume
- $w_i$: molecular weight of monomer of species
- $L$: length of time step
- $N$: number of grid points
- $P$: number of time stages
- $Q$: pass number
- $r$: region size used for allowable control
- $R$: number of allowable values for control used at each
- $t_f$: residence time or batch time
- $x_i$: state variable
- $x$: state vector
- $\gamma$: region contraction factor
- $\eta$: region restoration factor
- $\phi$: molar ratio of monomer
- $\phi_f$: monomer mol ratio in feed stream
- $\phi_i$: desired value of molar ratio of monomer in reaction mixture
- $\phi_t$: cross termination factor

References