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## Optimization of a sequence of reactors

by

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In this paper we are dealing with the optimal production of sulphuric acid in a sequence of reactors. Using a suitable approximation to the objective function, this problem can easily be solved using the maximum principle. A numerical example documents the applicability of the suggested approach.

### 1. Introduction

The case we are solving in this paper is taken from the theory of chemical reactors, but is of so elementary a character that it will be understandable without any prior knowledge of chemical engineering. Further details on the technical aspects of the problem can be found in Aris [1]. Here we are primarily dealing with the problem of optimizing a sequence of reactors. These problems have been discussed using dynamic programming in Aris [2] and the discrete version of the maximum principle in Fan and Wang [3].

In this paper we are specifically dealing with the production of sulphuric acid. It is known that high extent of reaction is obtained at a low rate of reaction,

therefore to obtain faster production the products are reacting in a sequence of continuous-flow reactors and by letting the temperature fall from reactor to reactor it is possible to achieve that the products react faster in the first reactor and slower in the last reactors. Obviously the optimal solution will demand an infinite number of reactors, but due to the investment cost the number of reactors is usually reduced to 3 – 5 reactors.

In Section 2, our problem will be formulated as a discrete-time control problem. In Section 3, the optimality conditions based on the discrete version of the maximum principle will be formulated and it is also shown that this system of equations can be easily solved. Section 4 presents the numerical solution of the case in study, and finally the last section gives some final remarks.

## 2. The optimization problem

The case in study is the production of sulphuric acid in a series of  $N$  reactors, that in our case has been fixed to  $N = 4$ . The whole process is illustrated below:

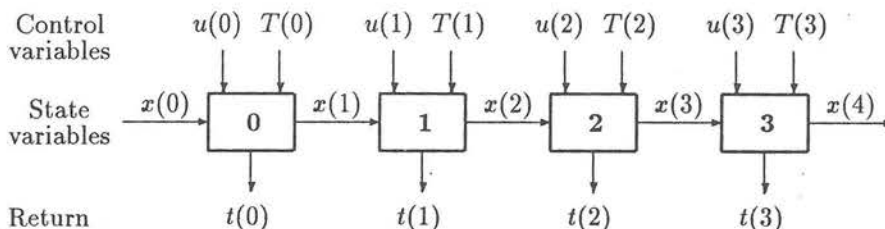


Figure 1. The discrete-time control problem.

where the *state* variables are  $x(i + 1)$ , the extent of reaction, and measure how far reaction has been progressed in stage  $i$ . Then  $x(i + 1) = x(i) + u(i)$ ,  $i = 0, 1, \dots, N - 1$ , where  $u(i)$  is the increase of the extent of reaction at period  $i$ . The control variables are  $u(i)$  and  $T(i)$  where the last one is the temperature at the  $i^{\text{th}}$  stage.

Moreover we have the relation for the holding time:

$$t(i) = \frac{u(i)}{r(x(i) + u(i), T(i))} \quad \begin{array}{l} \text{(gram mole/lit.)} \\ \text{(gram mole/lit./sec.)} \end{array} \quad (1)$$

where  $r()$  is the rate of reaction. The problem is to find the optimal control that minimizes the total time, and changes  $x()$  from  $x(0) = 0$  to  $x(N) = a$ , a specified value less than one.

Formally our problem is then

$$F = \min_{\{u(i), T(i)\}} \left[ \sum_{i=0}^{N-1} \frac{u(i)}{r(x(i) + u(i), T(i))} \right] \quad (2)$$

subject to

$$x(i+1) = x(i) + u(i), \quad \text{for } i = 0, 1, \dots, N-1 \quad (3)$$

$$\begin{aligned} x(0) &= 0 \\ x(N) &= a \end{aligned} \quad (4)$$

$$u(i) \geq 0, \quad \text{for } i = 0, 1, \dots, N-1 \quad (5)$$

As the  $T(i)$ 's are independent of each other (2) can be expressed as:

$$F = \min_{\{u(i)\}} \left[ \sum_{i=0}^{N-1} \frac{u(i)}{\max_{T(i)} r(x(i) + u(i), T(i))} \right] \quad (6)$$

Now for given values of  $x(i) + u(i)$ , the values of  $T(i)$  that minimizes  $r()$  can be uniquely determined using a one-variable search procedure. In this way, we find the function  $R()$ :

$$R(x(i+1)) = \max_{T(i)} r(x(i) + u(i), T(i)) \quad (7)$$

That is graphically illustrated in Figure 2.

The function  $R()$  for our actual case in study is in Figure 3. We will now approximate this function by the following expression

$$R(x(i+1)) = k_1 e^{-k_2(x(i)+u(i))}, \quad k_1, k_2 > 0 \quad (8)$$

This approximation is also shown in Figure 3.

Note that this approximation is best in the tail of the function  $R()$ , this will be sufficient for our practical purposes because as shown in Section 4 the optimal  $x(i)$ 's will be located at the tail of the function  $R()$ . Moreover, by varying  $k_1$ ,  $k_2$  different approximations could be obtained, (in the last section we will continue this discussion).

Obviously, for cases where the  $x(i)$ 's will take small values it will be better to approximate the function  $R()$  by a two-exponential curve of the form

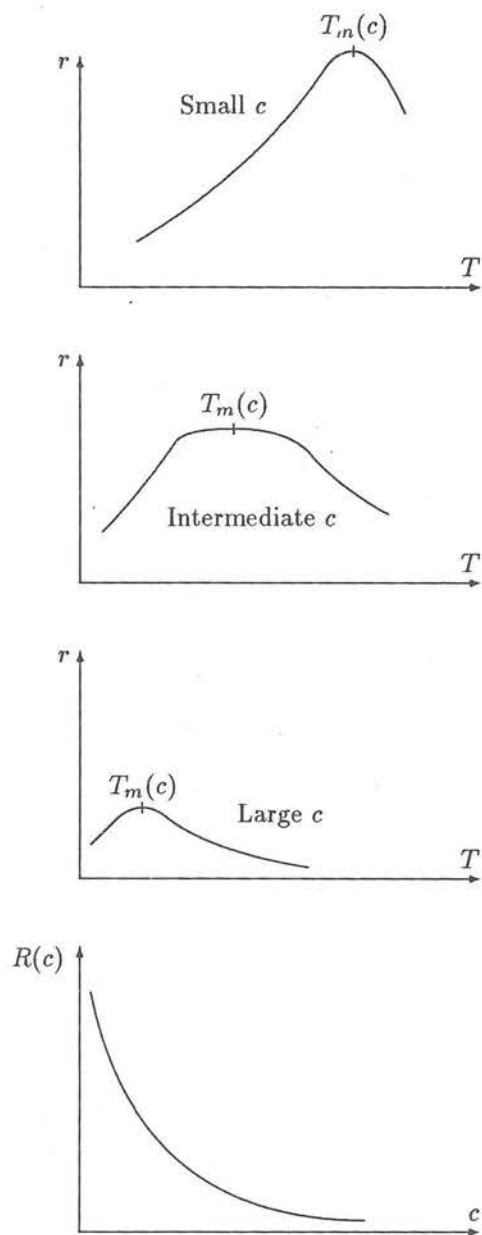


Figure 2. The rate of reaction functions  $r(c, T)$  and  $R(c)$ , where  $T$  is temperature in  $^{\circ}\text{K}$  and  $c$  is the extent of reaction in (gram mole/lit.).

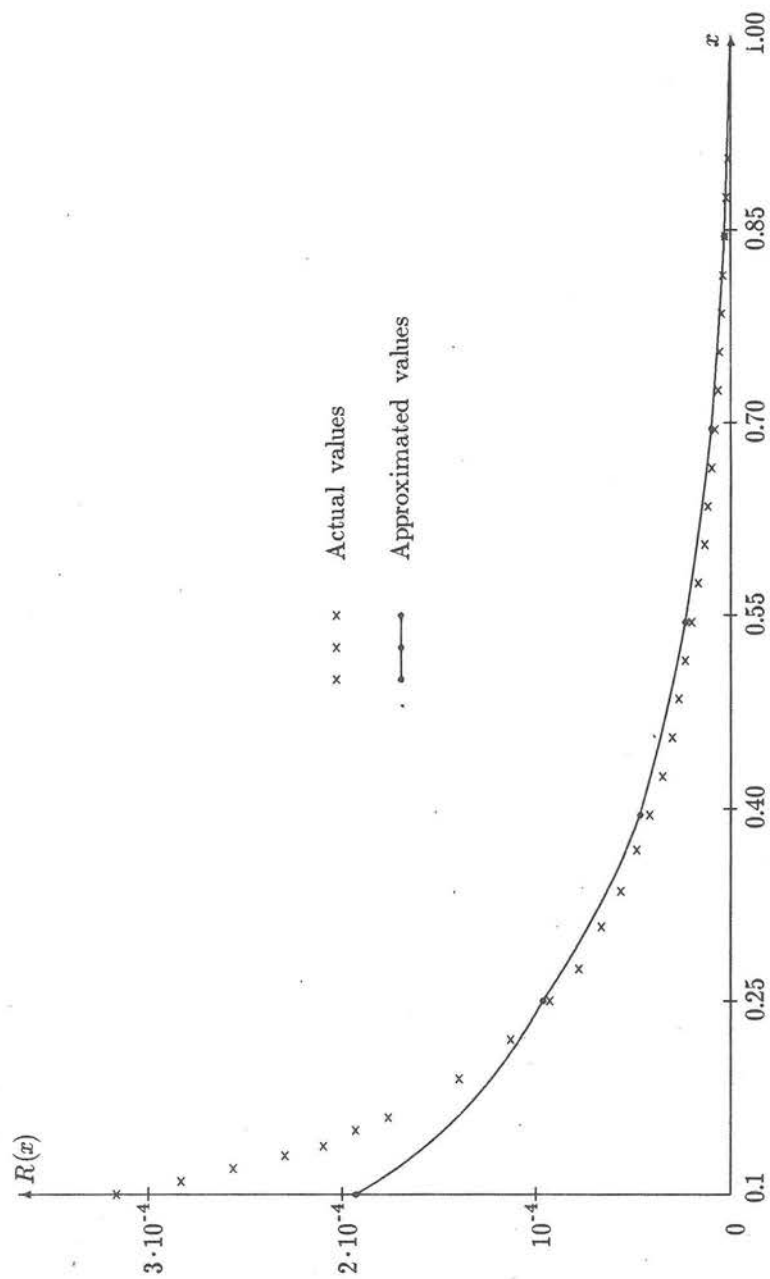


Figure 3. Maximum rate of reaction as a function of extent of reaction.

$$R(x(i+1)) = k_1 e^{-k_2 x(i+1)} + k_3 e^{-k_4 x(i+1)}$$

In such a situation the analytical approach of Section 3 is still valid but the number of parameters to be estimated is doubled.

Although the optimization of (6) could be directly performed on the original function, this approximation will permit us to develop an elegant way to find the optimal solution as it will be shown in the next section. The elegance of the method resides in the fact that we obtain quasi-analytical results due to the fact, as it will be shown later, that in the region of interest the criterion is strictly concave.

Finally, note also that the optimal solution  $\{u^*\}$  is dependent of  $k_1$ .

### 3. The optimality conditions

Our problem is to find the optimal solution of the following discrete-time problem:

$$k_1 F = \min_{\{u(i)\}} \sum_{i=0}^{N-1} u(i) e^{k_2(x(i)+u(i))} \quad (9)$$

subject to

$$x(i+1) = x(i) + u(i), \quad i = 0, 1, \dots, N-1 \quad (10)$$

$$x(0) = 0 \quad (11)$$

$$x(N) = a, \quad \text{and} \quad k_2 > 0 \quad (12)$$

$$u(i) \geq 0 \quad (13)$$

The necessary conditions for the optimality of  $x(i)$  and  $u(i)$  are given by the following relations (Vidal [4]):

$$\max_{u(i) \geq 0} \mathcal{H}(x^*(i), u(i), p^*(i+1)), \quad \text{for} \quad i = 0, 1, \dots, N-1 \quad (14)$$

$$\frac{\partial \mathcal{H}}{\partial x^*(i)} = p^*(i), \quad \text{for} \quad i = 0, 1, \dots, N-1 \quad (15)$$

$$\frac{\partial \mathcal{H}}{\partial p^*(i+1)} = x^*(i+1), \quad \text{for} \quad i = 0, 1, \dots, N-1 \quad (16)$$

and the boundary conditions:

$$\begin{aligned} x(0) &= 0 \\ x(N) &= a \end{aligned} \quad (17)$$

where  $\{p(i)\}$  are the adjoint variables and the Hamiltonian is defined as:

$$\mathcal{H}(x(i), u(i), p(i+1)) = -u(i)e^{k_2(x(i)+u(i))} + p(i+1)(x(i) + u(i)) \quad (18)$$

It is easy to show that the Hamiltonian is quasiconcave in  $u(i)$  (actually for  $u(i) \geq 0$ ,  $\mathcal{H}$  is strictly concave). Moreover for given values of  $x(i)$ ,  $p(i+1)$ , the Hamiltonian has a unique, bounded maximum that is achieved at a finite value  $u(i) \geq 0$ . Therefore the necessary conditions give also sufficient conditions for optimality, i.e. the directional convexity of the set of extended states' assumption is satisfied, see further Nahorski et al [5].

Assume now that at the optimal solution  $u^*(i) \neq 0$ , then (14) necessarily becomes:

$$\frac{\partial \mathcal{H}}{\partial u(i)} = 0 \quad (19)$$

or:

$$-e^{k_2(x(i)+u(i))} - k_2 u(i) e^{k_2(x(i)+u(i))} + p(i+1) = 0 \quad (20)$$

for  $i = 0, 1, \dots, N-1$ .

And the adjoint equations become:

$$-k_2 u(i) e^{k_2(x(i)+u(i))} + p(i+1) = p(i) \quad (21)$$

Comparing (20) and (21), we find the following conditions:

$$p(i) = e^{k_2(x(i)+u(i))} \quad (22)$$

for  $i = 0, 1, \dots, N-1$ , from where we obtain:

$$u(i) = \frac{\ln p(i)}{k_2} - x(i), \quad \text{for } i = 0, 1, \dots, N-1. \quad (23)$$

$$x(i) = \frac{\ln p(i-1)}{k_2}, \quad \text{for } i = 1, \dots, N. \quad (24)$$

Replacing (23) and (24) in (21), we obtain the following system of equations in  $\{p(i)\}$ :

$$\ln p(N-1) = k_2 a \quad (25)$$

$$\frac{p(i+1)}{p(i)} = 1 + \ln \frac{p(i)}{p(i-1)}, \quad \text{for } i = 1, \dots, N-2 \quad (26)$$

$$\frac{p(1)}{p(0)} = 1 + \ln p(0) \quad (27)$$

Let us now reduce this system of equations to a single equation in  $p(0)$ , by introducing the following operators:

$$L_0(x) = \ln(ex) \quad (28)$$

$$L_{i+1}(x) = \ln(eL_i(x)), \quad \text{for } i = 0, 1, \dots, N-3 \quad (29)$$

Then it is easily shown that

$$p(i+1) = p(0) \cdot \prod_{j=0}^i L_j(p(0)), \quad \text{for } i = 0, 1, \dots, N-2. \quad (30)$$

Thus we obtain one single equation on  $p(0)$ :

$$p(0) \cdot \prod_{j=0}^{N-2} L_j(p(0)) = e^{k_2 a}, \quad (31)$$

that can be solved by any iterative method. Thereafter it is easy to calculate the other  $p(i)$ 's and the optimal  $x(i)$ 's and  $u(i)$ 's.

#### 4. An example

Let us illustrate our approach by solving the case illustrated in Section 2, where

$$k_2 = 4.79$$

$$k_1 = 3.17 \cdot 10^{-4}$$

$$a = 0.9700$$

Solving equation (31) gives

$$p^*(0) = 8,680$$

and thereafter we can calculate from (30)

$$p^*(1) = p(0) \ln(ep(0)) = 27.438$$

$$p^*(2) = 59.016$$

$$p^*(3) = 104.214$$

And the optimal values of control and state variables are calculated from (23) and (24)

$$u^*(0) = 0.4512 \quad x^*(1) = u^*(0)$$



$$\begin{aligned}
 u^*(1) &= 0.2403 & x^*(2) &= 0.6914 \\
 u^*(2) &= 0.1599 & x^*(3) &= 0.8513 \\
 u^*(3) &= 0.1187 & x^*(4) &= 0.9700
 \end{aligned}$$

and the value of the objective function is

$$F = \frac{1}{k_1} \sum_{i=0}^{N-1} u^*(i) \cdot p^*(i) = 1.011945 \cdot 10^5 \text{ sec.} \quad (32)$$

## 5. Final remarks

The calculations made before finding the optimal  $p^*(0)$  can be utilized to perform sensitivity analysis on the parameters  $a$  and  $k_2$ , by keeping one of them constant and varying the other. This is shown in table 1.

$p^*(0)$	$k_2 = 4.79$	$k_2 = 4.79$	$a = 0.97$	$a = 0.97$
	$a$	$k_1 F$	$k_2$	$k_1 F$
8.00	0.9435	28.3161	4.6593	29.0978
⋮	⋮	⋮	⋮	⋮
8.63	0.9682	32.0128	4.7809	32.0735
8.64	0.9685	32.0735	4.7828	32.1219
8.65	0.9689	32.1342	4.7846	32.1702
8.66	0.9693	32.1949	4.7865	32.2187
8.67	0.9697	32.2557	4.7883	32.2670
8.68	0.9700	32.3165	4.7902	32.3154
8.69	0.9704	32.3773	4.7920	32.3639
8.70	0.9708	32.4382	4.7938	32.4123
8.71	0.9711	32.4991	4.7957	32.4608
8.72	0.9715	32.5600	4.7975	32.5092
8.73	0.9719	32.6210	4.7993	32.5577
⋮	⋮	⋮	⋮	⋮
8.99	0.9813	34.2171	4.8460	33.8219

Table 1. Sensitivity analysis

The second and third column in Table 1, show the changes in  $F$  due to small changes in  $a$ , this information can be used to evaluate to what extent it pays to

modify the value of  $a = 0.97$ .

The last two columns show the changes in  $F$  due to small changes in  $k_2$ . Since  $k_2$  is an estimated value, this information can be used to find better value of  $k_2$  by comparing  $F$  with the actual criteria function, for a given  $k_1$ . This information can be used in another model with the purpose of finding "best" values of  $k_1$  and  $k_2$ .

## References

- [1] ARIS R., The Optimal Design of Chemical Reactors, New York, Academic Press, 1961.
- [2] ARIS R., Discrete Dynamic Programming, New York, Blaisdell, 1964.
- [3] FAN L.T., WANG CH.S., The Discrete Maximum Principle, New York, Wiley, 1964.
- [4] VIDAL R.V.V., Notes on Static and Dynamic Optimization, IMSOR, The Technical University of Denmark, Lyngby, 1981.
- [5] NAHORSKI Z., RAVN H., VIDAL R.V.V., The discrete maximum principle - a survey and some new results, *International Journal of Control* 40, (1984), 3, 533-554.

## Optymalizacja ciągu reaktorów

W pracy rozważono optymalizację produkcji kwasu siarkowego w ciągu reaktorów. Po zastosowaniu odpowiedniej aproksymacji wskaźnika jakości problem ten można łatwo rozwiązać za pomocą zasady maksimum. Przykład numeryczny potwierdza możliwości zastosowania proponowanego podejścia.

## Оптимизация технологической линии реакторов

В работе рассматривается оптимизация производства серной кислоты в технологической линии реакторов. После применения соответствующей аппроксимации показателя качества эту задачу легко решить с помощью принципа максимума. Численный пример подтверждает возможности применения предлагаемого подхода.