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Simulating the mass balances of a lactulose pilot plant reactor system ☆

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Simulating the mass balances of a lactulose pilot plant reactor system [☆]

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Abstract

A model was developed for the reaction kinetics for converting lactose to lactulose using a sodium hydroxide catalyst and a boric acid/lactulose complexation reagent. Using the model, a reactor system was developed which consisted of two continuous stirred tank reactors followed by a tubular reactor. We simulated the reactor system using the ERRC Food Process Simulator by replacing one of the subroutines in the prototype program with a model specific for the reaction kinetics. The simulation accurately calculated the mass balances for the various streams associated with the three reactors.

Keywords: Simulate; Lactulose; React

1. Introduction

Process simulators have become fairly common in many engineering disciplines. However, their introduction to the food industry is quite recent. The primary needs for a food engineer are material and energy balances. An exact simulation of the process is not an absolute requirement [1].

Frequently, the simulators for the food industry are specific for a particular commodity or were adapted to a food process from an unrelated field. Consequently, most simulators are somewhat unwieldy and restricted to a narrow choice of food processes.

We set out to develop a simulator which would be portable to all food processes [6]. As a prototype we developed a simulator which, while specific to one process, potato flakes, is readily portable to other processes. It is open-ended. A user can supply models relevant to their process, theoretically based or empirical. The Fortran

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source code is available at no charge to domestic users. Anyone with a knowledge of engineering and Fortran can adapt it to a different process. It runs on a PC and is IBM compatible.

To demonstrate the portability of the ERRC Food Process Simulator to processes other than potato processing we took advantage of a continuous pilot plant process under development at our Center for making lactulose from surplus lactose. Lactulose is a high cost pharmaceutical with extensive markets in Japan and Europe. It is normally made by isomerizing lactose in the presence of base giving low yields with multiple side products. We used a boric acid selective complexation reagent to achieve yields of about 75% [2].

The reaction model consists of two consecutive but separate reactions [3]. In the first, lactose isomerizes to lactulose and complexes with boric acid. The formation of a complex shifts the equilibrium in favor of lactulose. Acidification breaks the complex releasing the lactulose. We have developed the pilot plant system of reactors for the lactulose process, but not the separation and recovery sections. Research is continuing on this phase. Our objective in this study was to demonstrate the portability of the ERRC Food Process Simulator to processes other than food processes by development of a simple procedure for adapting the simulator to the lactulose reaction system.

2. Modelling and simulation environment

2.1. Continuous pilot plant reactor process

Fig. 1 shows the reactor system consisting of three reactors in series. The first two were Continuous Stirred Tank Reactors (CSTR-A and CSTR-B) followed by a double pass tubular reactor (TR-C). The hold-up volume of each CSTR was 22.7 l. The volume of the tubular reactor was 2.6 l.

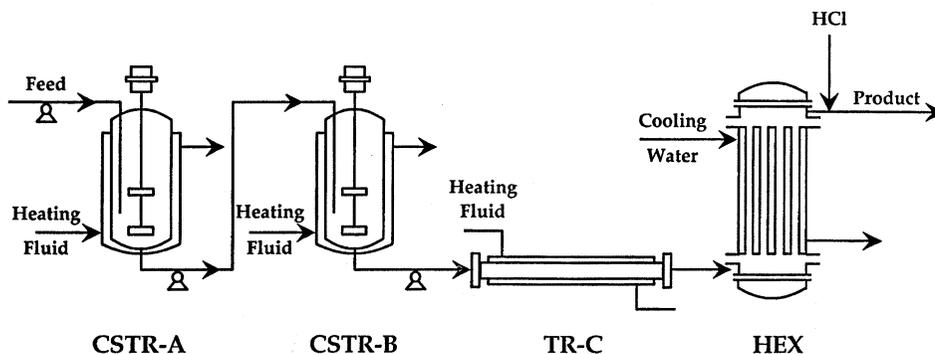


Fig. 1. Schematic of the continuous reactor system showing two continuous stirred tank reactors (CSTRs), a tubular reactor (TR) and a heat exchanger (HEX).

The effluent from the tubular reactor went to a sanitary plate and frame heat exchanger (HEX) to cool the product stream below room temperature.

Enough feed (140 l) was charged to the feed kettle to supply the reactors for approximately 4.5 h at a rate of 0.53 l/min.

2.2. Overview of the simulator program

The program consists of three sections: (1) a basic structure or program shell on which to build, (2) models, for each unit operation in a process, and (3) a readily understandable source code so other engineers could add to it and tailor it to other food processes. The main program is the interface to the user where data and a digital representation of the process are entered. This program calls the unit operations subroutines in the proper sequence. The main program includes two trial and error routines. One is an accelerated method and one is direct substitution. Although we developed the prototype based on the potato flake process, it can readily be used to simulate, for example, cheese manufacture, wine development, chocolate processing, or even pizza sauce preparation. After all, since the models for the different steps in the process are available, the computer does not know the difference between cookies and frozen peas.

2.3. Description of the main program

The main program or shell was written in three sections. The first section initializes the process. We wrote a small program just to initialize the input data files. The program uses the same data files to input and store the simulation data for the process.

The program uses two data files. Both are text files. One contains the variables associated with the individual streams which are specified either because they are known input streams or are streams which must be specified or guessed for a trial and error calculation. The other data file contains cost factors, coefficients for physical properties such as diffusivities, and the specifications and parameters for the unit operations in the process.

The heart of the shell is the second section. Here the engineer specifies the unit operations. In essence, it is here that the engineer gives the computer the flow sheet. Rather than an analog drawing of the flow sheet, this is a digital flow sheet – the type and sequence of unit operation are entered.

Each unit operation in the digital flow sheet accesses a subroutine which contains the algorithms for that unit operation. Initially, only potato process unit operations were present. Now we have added unit operations for non-food process unit operations.

The third section gives the answer; the material and energy balances are recorded in an answer data file which can be read interactively or printed. It is a text file.

2.4. Subroutines for unit operations models and utility functions

There are two levels of subroutines. The main program or shell accesses the first level, the subroutines for major unit operations. The second level, accessed through the first level subroutines, consists of the cost subroutine and auxiliary subroutines which are not unit operations, such as the trial and error subroutines. Data passes to the subroutines through the arguments in the CALL statements and through COMMON blocks, which are the input data files. Stream parameters, temperature, pressure, enthalpy, flow rate, and concentrations of components are stored in the data files and passed between the main program and the subroutines by the COMMON blocks. The identification of the streams and specific data for a particular unit operation are passed to the subroutines through the CALL statements. For the REACTOR subroutine, all streams are given initial values or assigned a value of 0.0 in the COMMON block. The CALL statement transfers the number for the streams, a factor for the fraction of the flow which is backmixed, dwell time, reactor size, and the kinetic model constants.

3. Results and discussion

3.1. Model development

We developed the kinetics model using bench scale reaction studies [3]. The rate controlling reaction is the isomerization of lactose to lactulose and the formation of a boric acid-lactulose complex. Therefore, the reaction kinetics were modeled as a first-order disappearance of lactose, Eq. (1).

$$dL/d\theta = -k_1L, \quad \text{where } k_1 = [8.389 \cdot 10^{18}] \exp[-16095/T]. \quad (1)$$

From a practical, engineering approach, we developed a model subject to boundary conditions such as pH and molar ratio of boric acid to lactose. As shown in Fig. 2 a molar ratio of slightly above 1.0 and a pH of 11 give the greatest yield. Fig. 3 shows a temperature range of 70–71°C gives the highest equilibrium conversion. Therefore, we modeled the kinetics at these conditions.

Integrating the kinetics model of Eq. (1), gives

$$L/L_0 = \exp(-k_1\tau), \quad \text{where } \tau = \text{reaction time and } k_1 = 0.041 \text{ min}^{-1} \text{ at } 71^\circ\text{C}. \quad (2)$$

The conversion to lactulose was found to obey Eq. (3),

$$ILS = ILS_{eq} [1 - \exp(-k_1\theta)], \quad \text{where } ILS_{eq} = 0.75. \quad (3)$$

3.2. Model validation

The continuous reactor system developed, Fig. 1, tested the accuracy of the model [4]. The process was set up at the optimum conditions indicated by the model;

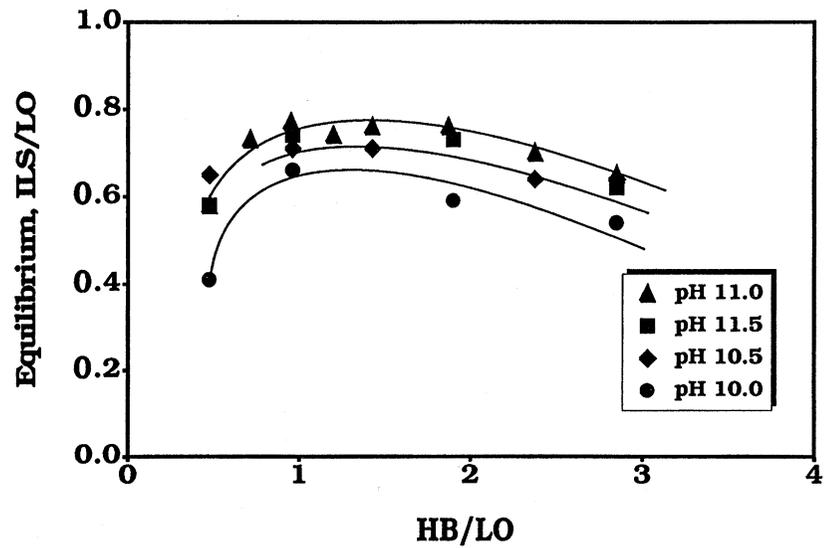


Fig. 2. Fractional conversion to lactulose as a function of molar ratio of boric acid to lactose at various pH levels. ILS/LO is the molar ratio lactulose/initial lactose conc. HB/LO is the molar ratio boric acid/initial lactose conc.

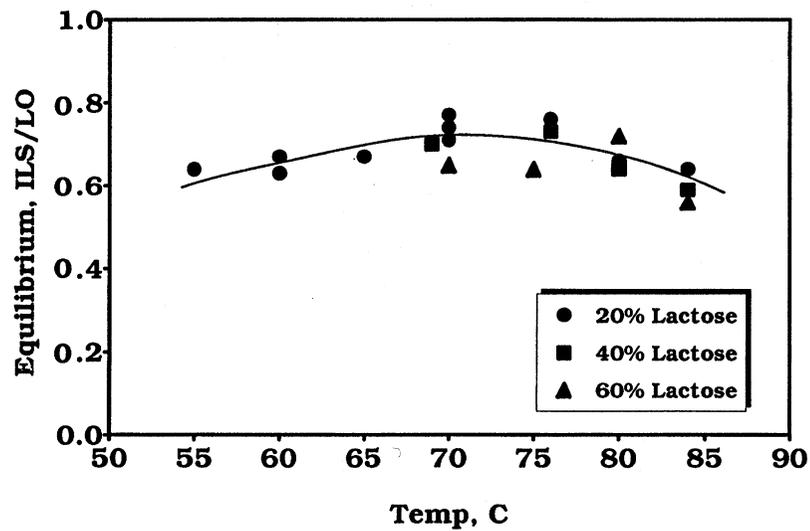


Fig. 3. Fractional conversion to lactulose as a function of reactor temperature. ILS/LO is the molar ratio lactulose/initial lactose conc.

namely, molar ratio just above 1.0, pH of 11, and a temperature of 70–71°C. The model predicts a batch reactor will approach equilibrium conversion of 75% in 88 min. Assuming perfect mixing in the two CSTRs with a nominal residence time

of 44 min each, the model predicts the conversion in a continuous flow process at steady state should be 66%. We added a short tubular reactor after the CSTRs. Assuming plug flow, the model predicts the final conversion to be 67%. Of course, mixing was not perfect backmix in the CSTRs or plug flow in the tubular reactor. The actual conversion for eleven experiments was an average of 76%. Further study of the reactor system indicated there was appreciable plug flow regime in the CSTRs.

3.3. Simulation results

We designed the program to permit adding new models by including a nonexecutable subroutine. This subroutine is to be used as an example for adding a subroutine. To add a model replace the prompts in main program for this subroutine and add the algorithm in the subroutine. The location for the algorithm is indicated within the code.

There is an alternate method for adding subroutines to a non-potato process. If the process does not use the unit operations in a potato process, simply substitute new algorithms into an existing subroutine. For example, if we were to simulate a cheese process, many of the unit operations would not be relevant. We might want to replace the hot water blancher for potatoes with the cheese inoculation tank – with the appropriate model. The hard part is the development of the model.

In this study we chose to use the substitution method. The original simulator program contains a subroutine called BLANCH which, in addition to calculating leaching due to diffusion, corrects the effluent from a hot water blancher for non-ideal flow [5]. The actual concentration of reactant and product in a flow reactor such as a CSTR is a function of mixing as well as reaction. This subroutine assumes two types of flow are present; backmix and plug flow. The inlet stream is split with part of the stream treated as backmix and part as plug flow. For a perfectly mixed (complete backmixing) flow reactor (CSTR) the concentration of product mixture in the effluent is a function of Eq. (4),

$$C/C_0 = \exp(-\theta/\theta_0), \quad \text{where } \theta/\theta_0 \text{ is the reduced dwell time,} \quad (4)$$

To correct the kinetics model for backmixing, we multiplied Eq. (3), the kinetics equation, by (1 – Eq. (4)), the correction for backmixing:

$$ILS = ILS_{eq} [1 - \exp(-k_1\theta)] * [1 - \exp(-\theta/\theta_0)]. \quad (5)$$

To calculate conversion in plug flow we used Eq. (3) with no correction.

The two split streams are then recombined as one outlet stream. The fraction as each stream, backmix or plug flow, is determined by trial and error from actual data and incorporated into the program as a correction factor. The correction factor is the fraction of the input stream treated as backmixed.

Our process data indicated the fractional conversion to lactulose leaving reactor A was 0.59–0.61, reactor B was 0.71–0.73, and reactor C was 0.73–0.75. Therefore we adjusted the correction factor (fraction backmixing) in each reactor to achieve this distribution. Using correction factors of 0.28, 0.60, and 0.10 respectively, the conversions were as listed in Table 1.

Table 1
Simulation results

Reactor	Correction factor ^a	Fraction conversion	
		Experimental	Simulation
A	0.28	0.59–0.61	0.60
B	0.60	0.71–0.73	0.73
C	0.10	0.73–0.75	0.74

^a Correction factor = fraction backmixed.

Fig. 4 shows how quickly the conversion drops off as the reactor temperature is lowered. Yet the simulation indicates the reactors should be run at a higher temperature. However, earlier work [3], Fig. 3, showed raising the temperature above about 71–73°C only lowers the yield due to degradative side reactions which limit the conversion. This contradiction exists because the kinetics model uses the Arrhenius equation with no boundary conditions and the side reactions, which are not fully known, are not included in the model. In simulating a specific reactor system such as this it is pragmatic to know and heed the limitations of the simulation rather than program in every boundary condition.

Fig. 5 shows the effect of residence time in the CSTRs. The tubular reactor residence time was unchanged. Note that the residence time used in our process,

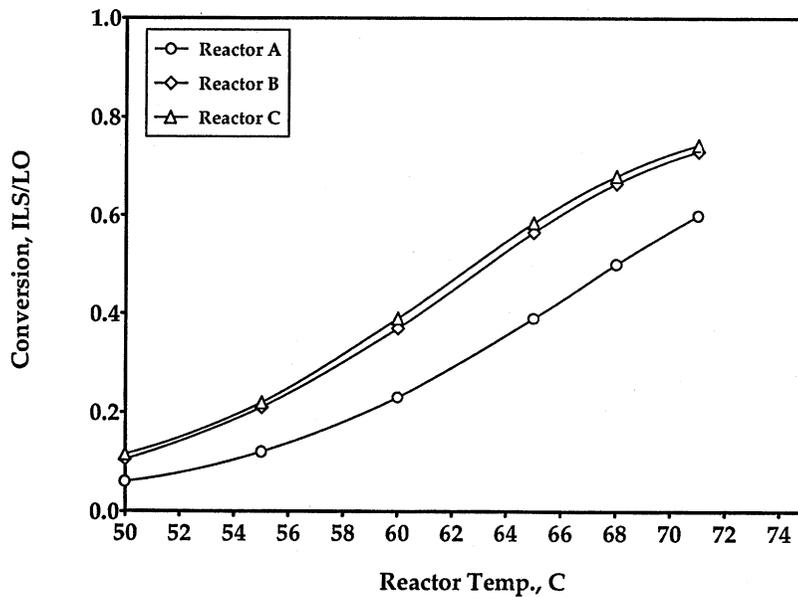


Fig. 4. Fractional conversion to lactulose in the reactor system as a function of reactor nominal temperature.

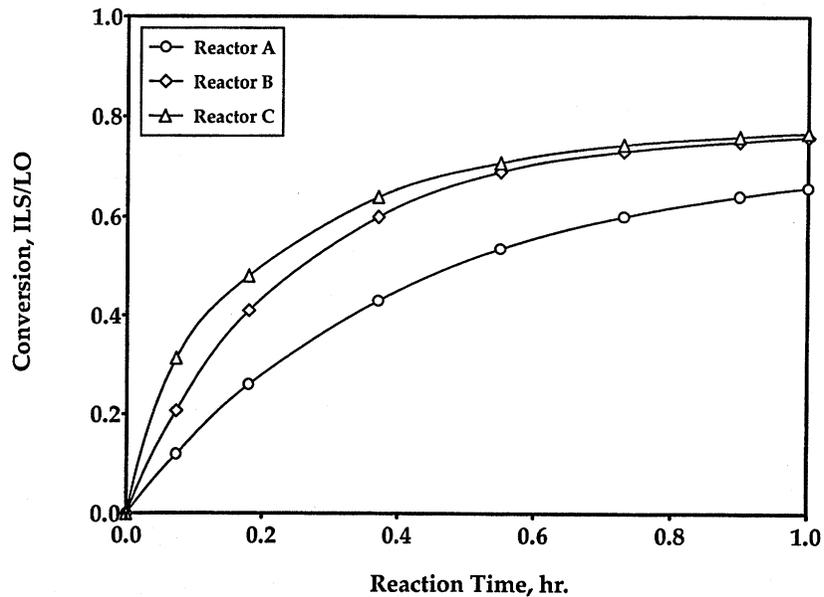


Fig. 5. Fractional conversion to lactulose in the reactor system as a function of reactor nominal dwell time in reactors A and B, 44 min each and 4.9 min in the tubular reactor.

0.73 h or 44 min, is just about at the asymptotic limit of conversion. The simulation confirms the choice of process conditions.

4. Conclusions

To expand the simulator program to other food processes involves two adjustments. The first adjustment is the writing of a new subroutine for each new unit operation. The second adjustment is adding new arguments to the main shell to input the appropriate unit operations parameters and open a path to the new subroutine.

For example, writing a new subroutine involves programming the equations which determine the mass and energy balances. The energy balance gives the temperature and enthalpy of all streams for the unit operation. The equations for mass transfer and conversion (reaction) give the mass flow rates and concentrations.

The second adjustment, adding arguments to the main program, is more complicated programming. The name for a unit operation, such as REACTOR, must be recognized by the computer and assigned a numerical designation corresponding to a subroutine. The program must prompt the user for each variable and parameter associated with that unit operation and correctly assign and transfer the values to the subroutine. Then, the main program must access the subroutine with its arguments corresponding to the proper sequence in the process flow sheet.

This particular simulation of the pilot plant lactulose reactor system is not intended as a general reactor model to be applied to other processes; but, is fairly specific to this particular type process, a first order reaction using one homogeneous feed stream and one homogeneous product stream. However, it establishes the portability to simulating other reactors, dependent only on the availability of a kinetics model.

Once we have a good simulation of the process we can use the simulator to investigate other options. In this particular case we have already maximized the conversion to lactulose based on the kinetics model. The full usefulness of the simulation will be realized when the entire process is developed and simulated.

This study demonstrated that the ERRC Food Process Simulator is portable to processes other than potato processes. We demonstrated the technique of substituting new algorithms into an existing subroutine in the simulator. Using this technique it is relatively easy to expand the program to a new process. Most likely, future portability will be to other food processes.

Appendix. Notation

C	concentration
C_0	concentration into first reactor in series
L	lactose, lactose concentration
L_0	initial lactose concentration
k_1	rate constant for disappearance of lactose, min^{-1}
T	Absolute temperature, $^{\circ}\text{K}$
ILS	lactulose, lactulose concentration
θ	time, min
θ_0	volumetric reactor holdup/flow rate, min
τ	reaction time, dwell time, min

References

- [1] S. Chall, *Food Engineering* (February 1993) 54.
- [2] K.B. Hicks and F.W. Parrish, *Carbohydr. Res.* **82** (1980) 393–397.
- [3] M. Kozempel and M.J. Kurantz, *Chem. Tech. Biotech.* **59** (1) (1994) 25–29.
- [4] M. Kozempel and M.J. Kurantz, *Chem. Tech. Biotech.* **59** (3) (1994) 265–269.
- [5] M.F. Kozempel, J.F. Sullivan and J.C. Craig Jr, *Amer. Potato J.* **62** (1985) 69–82.
- [6] M.F. Kozempel, P.M. Tomasula and J.C. Craig Jr, *Simulation Practice and Theory* **2** (4–5) (1995) 221–236.

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