# MODAL DECOMPOSITION OF A NONLINEAR TUBULAR REACTOR MODEL : A CONTROL PERSPECTIVE

Jennifer Brown \* Denis Dochain \*\* Michel Perrier \*\*\* and Fraser Forbes \*

\* Department of Chemical and Materials Engineering, 536 Chemical and Materials Engineering Building, Edmonton, Alberta, Canada T6G 2G6 \*\* Cesame, Université Catholique de Louvain, av. G. Lemaître

4-6, 1348 Louvain-La-Neuve, Belgium (corresponding author) e-mail : dochain@csam.ucl.ac.be, fax : 32-10-472180

\*\*\* Département de Génie Chimique, Ecole Polytechnique de Montréal, Case postale 6079 succursale "Centre Ville" Montréal H3C 3A7, Canada

Abstract: This paper focuses on the application of modal decomposition to a nonlinear convection-reaction-diffusion distributed parameter system (DPS). More precisely it will concentrate on the dynamical model of an industrial pulp bleaching tubular reactor described by nonlinear partial differential equations (PDEs). The objective of the modal decomposition is to generate, for control design, a discretized finite-dimensional model that contains the dominant modes of the process dynamics. The modal decomposition has been performed on a linearized tangent model of the process. It results in eigenfunctions that are modified Bessel functions of complex order.

Keywords: Tubular reactor, distributed parameter systems, modal decomposition, chemical process, pulp bleaching.

## 1. INTRODUCTION

There are many industrial processes in which the states, outputs and control variables vary spatially as well and temporally. These processes are known as distributed parameter systems (DPS). The natural form of the models that describe distributed parameter systems are partial differential equations (PDEs), integral equations or trandscendental transfer functions (Ray, 1981). Currently, for simplification and control purposes, most industrial processes are represented by lumped parameter models, which are characterized by ordinary differential equations. However, a large number of these processes are actually distributed in nature, and simple lumped parameter models that ignore the spatially varying nature of the DPS will often

suffer from strong interactions and apparent time delays due to the underlying diffusion and convection phenomena inherent in these processes (Gay and Ray, 1995). Examples such as heat transfer and sheet forming processes, heat exchangers, reactors and bioreactors, are just a few of the many processes where the dependent variables may vary in space as well as in time. For the purpose of this work, we will concentrate on partial differential equation representations of DPS because they stem from fundamental momentum, energy and material balances for a process. More specifically, we will concentrate on one specific type of PDE: parabolic equations. Parabolic systems play an important role in the description of the dynamics of chemical processes. Parabolic equations can be

used to describe the dynamics of tubular reactors whenever dispersion phenomena are present. Typically parabolic equations modeling tubular reactors with axial dispersion can be viewed as a very general case, which is intermediate between the ideal cases: the continuous stirred tank reactor (CSTR) and the plug-flow reactor (PFR).

Research in the field of control of DPS has been ongoing since the 1970's. Ray published a survey of applications of distributed parameter systems theory, which encompassed a large number of fields, indicating a need for further applications research (Ray, 1978). Dochain (Dochain, 1994) notes that although research in the theory of DPS is still quite active, it faces two problems. The first is related to the high degree of abstraction of DPS: a lot of the research is done by mathematicians who handle rather abstract and complex mathematical objects (the mathematical complexity is due to the infinite dimensionality of DPS). This complexity then makes the connection with practical aspects of industrial process and systems very difficult. The second problem is that most results do not apply to nonlinear DPS (Dochain, 1994).

Analytical solution of partial differential equations describing DPS is generally nontrivial and in many cases impossible (Hanczyc and Palazoglu, 1995). Due to the mathematical complexity of PDEs, approximation methods have been studied to a great extent. Conventional approaches for control of DPS are based on spatial discretization of the PDE model, yielding a finite number of ordinary differential equations in time (Christofides and Daoutidis, 1996). The rich theory available for the control of lumped parameter systems can then be applied to the discretized system of ODEs. Unfortunately, common discretization techniques such as finite difference, finite element and finite volume methods often yield large systems of ODEs, making the problems computationally unattractive and losing information contained in the model. Also, notions such as controllability and observability may depend on the discretization method and the number and location of discretization points (Ray, 1981).

Efforts have been made to reduce the number of ODEs necessary to represent the true distributed parameter system. Orthogonal collocation and other weighted residual methods (Villadsen and Michelsen, 1978) can result in systems of lower order. For parabolic systems, modal decomposition has often been employed to provide approximate solutions and theoretical results regarding control of DPS (Gay and Ray, 1995). Murray-Lasso (Murray-Lasso, 1965) first presented modal decomposition for DPS in the 1960's, and it was developed to a great extent by Ray (Ray, 1981).

It is based on the ability to represent the spatially varying input and output of the system as a sum of an infinite series of the system spatial eigenfunctions and time-dependent coefficients. This is similar to the collocation and weighted residual techniques; however modal analysis has the distinct property that the spatial eigenmode coefficients are decoupled. Motivated by the fact that modal decomposition uses the natural structure of the PDE to solve for the spatial eigenfunctions (which is property that the above methods do not have), the purpose of this research is to investigate modal decomposition techniques and evaluate them for use in control applications.

This paper focuses on the application of modal decomposition to a nonlinear convection-reactiondiffusion distributed parameter system, i.e. the dynamical model of an industrial pulp bleaching tubular reactor. The major challenges encountered in this case study are two fold: first, the bleaching reactor model consists of a coupled set on nonlinear PDEs. These are first linearized resulting in a set of nonlinear ODEs which has no analytical solution. Numerical solution of these ODEs is followed by a step whereby functions were fitted to the steady state solutions. Modal decomposition of this linearized model involved the solution of the spatial ODE which involves four variable changes in order to obtain an ODE which has an analytical solution yielding the eigenfunctions. The resulting eigenfunctions of the spatial operator are Modified Bessel Functions of complex order, which are not common in the literature. A number of simulations as well as a comparative study with finite difference results was done.

# 2. DYNAMICAL MODEL OF THE PULP BLEACHING TUBULAR REACTOR

The principle objective of pulp bleaching is to achieve a high brightness. This objective must be met without compromising the strength of the final product, which can occur if there is significant cellulose degradation during bleaching. The color in the pulp is due for the most part to lignin: a natural polymer occurring in the pulp. In Kraft pulping (which is the method we use here), the bleaching is an extension of the fiber delignification process started in the digester. Chlorine dioxide is one of the most important chemicals used in bleaching chemical pulp because it reacts readily with lignin, yet does not react to a significant extent with the carbohydrates (cellulose) (Dence and Reeve, 1996).

The bleaching reactor model consists of a set of nonlinear coupled PDEs. The two reactants in the model are chlorine dioxide (C) and lignin (L).

The reaction term is a bilinear term. The PDEs describing the reactor dynamics are:

$$\frac{\partial L}{\partial t} = -v\frac{\partial L}{\partial z} + D\frac{\partial^2 L}{\partial z^2} - k_l L C \tag{1}$$

$$\frac{\partial C}{\partial t} = -v\frac{\partial C}{\partial z} + D\frac{\partial^2 C}{\partial z^2} - k_c LC \qquad (2)$$

and the boundary conditions are:

$$\frac{\partial L(0,t)}{\partial z} = \frac{v}{D} (L(0,t) - L_{in} - L_o)$$
(3)

$$\frac{\partial L(H,t)}{\partial z} = 0 \tag{4}$$

$$\frac{\partial C(0,t)}{\partial z} = \frac{v}{D} (C(0,t) - C_{in} - C_o) \qquad (5)$$

$$\frac{\partial C(H,t)}{\partial z} = 0 \tag{6}$$

where  $C_{in}$ ,  $L_{in}$ , H,  $C_o$  and  $L_o$  are the inlet chlorine and lignin concentrations, the bleaching tower height, and constant adjustment parameters determined from the kinetic studies done by (Savoie and Tessier, 1997).

#### 3. LINEARIZED TANGENT MODEL

The bleaching reactor model consists of a set of nonlinear coupled PDEs. This creates a problem, since modal decomposition is only possible on linear systems (Ray, 1981). The model must therefore first be linearized around a chosen operating trajectory.

The steady-state values of C and L,  $C_{ss}$  and  $L_{ss}$ , are solutions of the following differential equations:

$$-v\frac{dL_{ss}}{dz} + D\frac{d^2L_{ss}}{dz^2} - k_l L_{ss} C_{ss} = 0$$
 (7)

$$-v\frac{dC_{ss}}{dz} + D\frac{d^2C_{ss}}{dz^2} - k_c L_{ss}C_{ss} = 0 \qquad (8)$$

The ODEs (7)(8) are nonlinear and have no analytical solution. They must therefore be solved numerically (with the extra difficulty that the ODEs have *initial and final* conditions). The steady state profiles of both variables have been computed for the following parameter values of the industrial pulp bleaching reactor :

$$k_c = 0.006, \ k_l = 0.035, \ Lin_{ss} = 31 \ Kappa$$
  
 $Lo = 9 \ Kappa, \ Cin_{ss} = 2.5 \ g/l, \ Co = 1.3 \ g/l$   
 $v = 1/30 \ min, \ D = 0.5/30$ 

An exponential model was then fitted to the results of the numerical integration. Figure 1 shows the results of the integration. A least squares fit was done on the log of the data. Other trial



Fig. 1. Exponential fit of the steady-state profile

functions (e.g. polynomials of various orders) were tested to see if they resulted in better fit, however the exponential function provided the best fit. Also, the benefit of having chosen a simple exponential function to fit  $C_{ss}$  and  $L_{ss}$ , is that as a consequence, there exists an analytical solution of the spatial ODE. The resulting exponential models for both  $C_{ss}$  and  $L_{ss}$  are:

$$C_{ss} = 0.6459e^{-1.5675z} \tag{9}$$

$$L_{ss} = \frac{\xi_{in} + k_l C_{ss}}{k_c} = 15 + 3.7678e^{-1.5675z} (10)$$

If we let  $\tilde{C} = C - C_{ss}$  and  $\tilde{L} = L - L_{ss}$ , the linearized tangent model can be written as follows:

$$\frac{\partial \widetilde{L}}{\partial t} = -v \frac{\partial \widetilde{L}}{\partial z} + D \frac{\partial^2 \widetilde{L}}{\partial z^2} - k_l L_{ss} \widetilde{C} - k_l C_{ss} \widetilde{L} (11)$$
$$\frac{\partial \widetilde{C}}{\partial t} = -v \frac{\partial \widetilde{C}}{\partial z} + D \frac{\partial^2 \widetilde{C}}{\partial z^2} - k_c L_{ss} \widetilde{C} - k_c C_{ss} \widetilde{L} (12)$$

$$\frac{\partial L(0,t)}{\partial z} = \frac{v}{D} (\widetilde{L}(0,t) - \widetilde{L}_{in})$$
(13)

$$\frac{\partial L(H,t)}{\partial z} = 0 \tag{14}$$

$$\frac{\partial \widetilde{C}(0,t)}{\partial z} = \frac{v}{D} (\widetilde{C}(0,t) - \widetilde{C}_{in})$$
(15)

$$\frac{\partial \widetilde{C}(H,t)}{\partial z} = 0 \tag{16}$$

#### 4. MODAL DECOMPOSITION

#### 4.1 Model triangularization

Modal decomposition is carried out on the linearized tangent model (11)-(16). Because of the coupling of the equations (11)(12), a first step then consists of introducing a state transformation (which corresponds to a system triangularization) so that the first equation becomes independent of the second one in order to allow to perform the modal decomposition in a recursive way. Let us consider the following state transformation :

$$\eta_1 = k_c \widetilde{L} - k_l \widetilde{C}, \ \eta_2 = \widetilde{C} \tag{17}$$

This change of variable yields the following set of PDEs with boundary conditions:

$$\frac{\partial \eta_1}{\partial t} = -v \frac{\partial \eta_1}{\partial z} + D \frac{\partial^2 \eta_1}{\partial z^2}$$
(18)
$$\frac{\partial \eta_2}{\partial t} = -v \frac{\partial \eta_2}{\partial z} + D \frac{\partial^2 \eta_2}{\partial z^2} - k_c L_{ss} \eta_2$$

$$-C_{ss} (\eta_1 + k_l \eta_2)$$
(19)

$$\frac{\partial \eta_i(0,t)}{\partial z} = \frac{v}{D} (\eta_i(0,t) - \eta_{i,in}), \quad i = 1, 2 \quad (20)$$

$$\frac{\partial \eta_i(H,t)}{\partial z} = 0, \quad i = 1,2 \tag{21}$$

This transformation eliminated the reaction term from the first PDE and is related to the notion of reaction invariants (Gavalas, 1968) (Bastin and Dochain, 1990).

## 4.2 Modal decomposition of $\eta_1(z,t)$

We can proceed with the modal decomposition for the first variable  $\eta_1(z, t)$ . The first step is to assume that the state variable  $\eta_1$  and the input variable  $\eta_{1in}$  can be written as follows :

$$\eta_1(z,t) = \sum_{n=1}^{\infty} p_{\psi n}(t)\psi_n(z)$$
 (22)

$$\delta(z)\eta_{1in}(t) = \sum_{n=0}^{\infty} q_n(t)\psi_n(z)$$
(23)

Substituting (22)(23) into Equation (18) yields a set of separable ODEs in time and space:

$$\frac{dp_{\psi n}(t)}{dt} + \lambda_{\psi} p_{\psi n}(t) = q_n(t) \qquad (24)$$

$$D\frac{d^2\psi_n}{dz^2} - v\frac{d\psi_n}{dz} + \lambda_{\psi n}\psi_n = 0$$
(25)

$$v\frac{d\psi_n(0)}{dz} + D\psi(0) = 0 \tag{26}$$

$$\frac{d\psi_n(1)}{dz} = 0 \tag{27}$$

The solution to (25) (using separation of variables) is:

$$\psi_n(z) = G_n e^{\frac{v}{2D}z} \left[ \cos\left(\frac{s_{\psi n}}{2D}z\right) + \frac{v}{s_n} e^{\frac{v}{2D}z} \sin\left(\frac{s_{\psi n}}{2D}z\right) \right] 28)$$

where:

$$s_{\psi n} = \sqrt{v^2 - 4D\lambda_{\psi n}} \tag{29}$$

On application of the boundary conditions,  $s_{\psi n}$ , are found by solving:

$$\tan\left(\frac{L}{2D}s_{\psi n}\right) = \frac{2vs_{\psi n}}{s_{\psi n}^2 - v^2} \tag{30}$$

Therefore the eigenvalues,  $\lambda_{\psi n}$ , can be found by rearranging (29):

$$\lambda_{\psi n} = -\frac{s_{\psi n}^2 + v^2}{4D} \tag{31}$$

Using the properties of orthogonal functions and Sturm-Liouville theory, the  $G_n$  coefficients are chosen such that  $\|\psi_n\|_2 = 1$ . With the eigenvalues,  $\lambda_{\psi n}$ , one may then solve (24). The solution is then given by:

$$\eta_{1}(z,t) = \sum_{n=1}^{\infty} \left[ e^{\lambda_{\psi n} t} \left\langle \eta_{1,0}, \widetilde{\psi_{n}} \right\rangle \psi_{n} + \int_{0}^{t} e^{\lambda_{\psi n} (t-s)} \left\langle \delta(z) v \eta_{1in}, \widetilde{\psi_{n}} \right\rangle \psi_{n} \right] (32)$$

where  $\widetilde{\psi_n}$  are the eigenfunctions of the adjoint operator (where  $M_n$  are chosen such that  $\left\|\widetilde{\psi_n}\right\|_2$ = 1) :  $\widetilde{\psi_n} = M_n \psi_n (L-z)$ .

# 4.3 Modal decomposition of $\eta_2(z,t)$

The modal decomposition of (19) is more complicated, due to the exponential terms in 'z', and the presence of a term in  $\eta_1$ . Substituting (9)(10) into (19) and combining terms yields:

$$\frac{\partial \eta_2}{\partial t} = -v \frac{\partial \eta_2}{\partial z} + D \frac{\partial^2 \eta_2}{\partial z^2} - \left(0.09 + 0.04521e^{-1.5675z}\right) \eta_2 - \left(0.6459e^{-1.5675z}\right) \eta_1$$
(33)

If the term in  $\eta_1 = 0$ , then (33) may be decomposed using the separation of variables method shown above. In that case, after separation of variables, an eigenvalue problem is formulated and can be solved. The eigenfunctions, together with appropriate time-dependent functions and some free constants, can then be combined in an infinite series (Street, 1973). Since  $\eta_1(z,t) \neq 0$ , one must solve the *associated* eigenvalue problem (with  $\eta_1$ = 0), with  $\eta_1$  regarded as an input. As before, we let:

$$\eta_2(z,t) = \sum_{n=0}^{\infty} r_n(t) \gamma_{1n}(z)$$
 (34)

We obtain ODEs in the spatial and time variables:

$$-v\frac{d\gamma_{1n}}{dz} + D\frac{d^{2}\gamma_{1n}}{\partial z^{2}} + \left[\lambda_{\gamma} - 0.09 - 0.04521e^{-1.5675z}\right]\gamma_{1n} = 0 \quad (35)$$
$$D\frac{d\gamma_{1n}(0)}{dz} - v\gamma_{1n}(0) = 0 \quad (36)$$

$$\frac{d\gamma_{1n}(1)}{dz} = 0 \quad (37)$$

$$\frac{dr_n}{dt} + \lambda_\gamma r_n = 0, \ r_n(0) = r_{n,0}(38)$$

The solution is based on a sequence of four successive change of variables, as suggested in (Murphy, 1960) (starting with Entry #106 in Tables of Equations of Second Order, p.321)(for further details, see (Brown, 2001)). This yields an ODE of the form:

$$\varpi_5^2 \frac{d^2 \gamma_{5n}}{d \varpi_5^2} + \varpi_5 \frac{d \gamma_{5n}}{d \varpi_5} + \left( \varpi_5^2 - p^2 \right) \gamma_{5n} = 0 \tag{39}$$

Equation (39) is the well known Bessel's Equation (Entry #274 in Murphy, p.331). Due to all the changes in variables and the numerical values of the constants, it is actually Bessel's modified equation (#275 in Murphy). The analytical solution to (35) is then given by ( $\alpha_1 = 1.5675$ ,  $\alpha_2 = 2.1015$ ):

$$\gamma_1(z) = \left(e^{-\alpha_1 z}\right)^{.6378} \\ \left[C_1 I_p \left(\alpha_2 \sqrt{e^{-\alpha_1 z}}\right) + C_2 I_{-p} \left(\alpha_2 \sqrt{e^{-\alpha_1 z}}\right)\right]$$

where  $I_p$  is a modified Bessel function of order pand argument  $\left(\alpha_2 \sqrt{e^{-\alpha_1 z}}\right)$ . Here, the order of the modified Bessel functions is given by:

$$p = \sqrt{10.4190 - 97.6778\lambda_{\gamma}} \tag{40}$$

We now proceed with the full solution using the boundary conditions given by (36)(37). This results in finding the values of  $\lambda_{\gamma n}$  such that (36)(37) are satisfied. The eigenvalues of the solution are found within the *order* of the modified Bessel function. It can be seen that upon application of the boundary conditions in this case, that the zeros of the Bessel functions will be functions of their orders (Gray and Mathews, 1931).

One other added complication is the value of (36)(37) are complex (non-real) for varying  $\lambda_{\dot{\gamma}n}$ . This makes finding the  $\lambda_{\gamma n}$ 's difficult, as root finding techniques for complex valued functions are not widely available. However, according to (Gray and Mathews, 1931), modified Bessel functions of the second kind  $K_p(z)$ , as well as the combined function  $I_p(az)K_p(bz) - I_p(bz)K_p(az)$ have no real zeros unless p is purely imaginary, and it can be shown that they have an infinite number of such zeros. Although (36)(37) is neither of the above functions, it was thought that there was a possibility that an infinite number of  $\lambda_{\gamma n}$ 's which satisfied the boundary condition did exist; especially since the order of the modified Bessel functions, p, in (36)(37) is purely complex.

The following procedure for finding the values of  $\lambda_{\gamma n}$  has been considered. Let us first denote the boundary conditions by B.C. = 0. As previously mentioned, the value of B.C. is not real valued, but is complex valued. Therefore in order to find where B.C. = 0, we use the fact that its

magnitude, |B.C.| should be zero when B.C. = 0. In other words, when  $B.C. = \alpha + i\beta$  with magnitude  $|B.C.| = \sqrt{\alpha^2 + \beta^2}$  and if B.C = 0, then: |B.C| = 0. One may then use conventional root solvers such as fsolve in Maple<sup>(C)</sup> to find the values of  $\lambda_{\gamma n}$  such that the boundary conditions are satisfied. The first five values of  $\lambda_{\gamma n}$  are equal to :  $\lambda_{\gamma 1} = 0.106666666$ ,  $\lambda_{\gamma 2} = 0.35581402$ ,  $\lambda_{\gamma 3} =$ 0.85265051,  $\lambda_{\gamma 4} = 1.6759089$ ,  $\lambda_{\gamma 5} = 2.8276587$ . Having found the first five eigenvalues, one can now express the full solution of  $\eta_2(z,t)$  using the eigenfunctions and the expression given by (34). Equation (41) shows in the first term, the "unforced" part of the solution; and the second term represents the "forced" part of the solution:

$$\eta_{2}(z,t) = \sum_{n=0}^{\infty} \left[ e^{\lambda_{\gamma n} t} \langle \eta_{2,0}, \widetilde{\gamma_{1n}} \rangle \gamma_{1n} + \int_{0}^{t} e^{\lambda_{\gamma n} (t-s)} \langle \eta_{1}(z,t), \widetilde{\gamma_{1n}} \rangle \gamma_{1n} ds \right] (41)$$

However in this case, one must note that although we consider only the unforced solution of  $\eta_2(z, t)$ , (i.e.  $\eta_{2,in}(t) = 0$ ), there is still a "forced" part which contains the input term in  $\eta_1(z, t)$ .



Fig. 2. Numerical simulation of  $\eta_1(z,t)$ 

### 5. SIMULATION RESULTS

Numerical simulations of both  $\eta_1$  and  $\eta_2$  have been performed. The simulation conditions were as follows : at t = 0, initial conditions were such that the reactor had a profile other than that of steady state ( $\eta_i(z, 0) = 0.1$ , i = 1, 2), and it was assumed that there was no variation from the steady-state value of the input ( $\eta_{i,in}(t) = 0$ , i = 1, 2). Figures 2 and 3 depict  $\eta_1(z, t)$  and  $\eta_2(z, t)$  from their initial states of 0.1 to the steady states with five modes. For comparison purposes a finite difference solution with two hundred spatial discretization points was done. A comparison was made by integrating the difference between the two solutions in space and in time (Figure 4). In general the error is fairly small (around  $10^{-4}$ ).



Fig. 3. Numerical simulation of  $\eta_2(z,t)$ 



Fig. 4. Integral of the difference between finite difference and modal decomposition

# 6. CONCLUSIONS

This paper has focused on the application of modal decomposition to a nonlinear convectionreaction-diffusion distributed parameter system, more precisely on the dynamical model of an industrial pulp bleaching tubular reactor described by nonlinear parabolic equations. The modal decomposition has been performed on a linearized tangent model of the process. It resulted in eigenfunctions that are modified Bessel functions of complex order. Since the objective of the modal decomposition is to generate, for control design, a discretized finite-dimensional model that contains the dominant modes of the process dynamics (Brown, 2001), particular care has been taken to perform in the most generic way so as to allow the application of the methodology to the largest possible class of nonlinear parabolic equations.

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