

FIXED TIME ZONE METHODOLOGY FOR PLUG FLOW SIMULATIONS AS APPLIED TO AN OXYGEN DELIGNIFICATION REACTOR, PART 2: THE PROCESS MODEL

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ABSTRACT

This is the second of a three part series of papers presented at the Control Systems 92 Conference at Whistler, B.C. describing the development of an advanced control application from concept to startup. Part 1, *High-Fidelity Real Time Dynamic Process Simulation with Object-Oriented Programming*, describes the tool and development environment while Part 3, *Advanced Control Development-Adaptive Nonlinear Control* describes the resulting control strategy.

This paper describes the process model used and introduces a fixed time zone model development methodology. Simulation studies showed the advantages of fixed time zone partitioning relative to conventional fixed position partitioning in the simulation of a plug flow process and demonstrated the high fidelity and flexibility achievable with the development tool utilized. Fixed time zone techniques were systematically applied for a high fidelity simulation of an oxygen delignification tower. The resultant model was applied for testing an advanced kappa number control scheme utilizing Simon's IDEAS™ simulation package.

INTRODUCTION

A high fidelity plug flow reactor model may validly be applied for both process design and control studies. The model must provide steady state accuracy and a satisfactory dynamic response to changing process inputs. In developing such a model the dynamics of both mixing and reaction kinetics must be characterized.

The conventional simulation of a plug flow process involves the partitioning of the vessel into a fixed number of spatial zones. The positions of the zone boundaries represent fixed surfaces through which transport equations define mass and enthalpy flows into and out of the fixed volumes of the zones. A serious difficulty arises with this approach in determining the interzonal flows in vertical pressurized vessels when gas and liquid phases coexist. Decisions made with respect to the spacing of the zones must be based on the range of volumetric flows into the vessel, the frequency range of process variable inputs, the

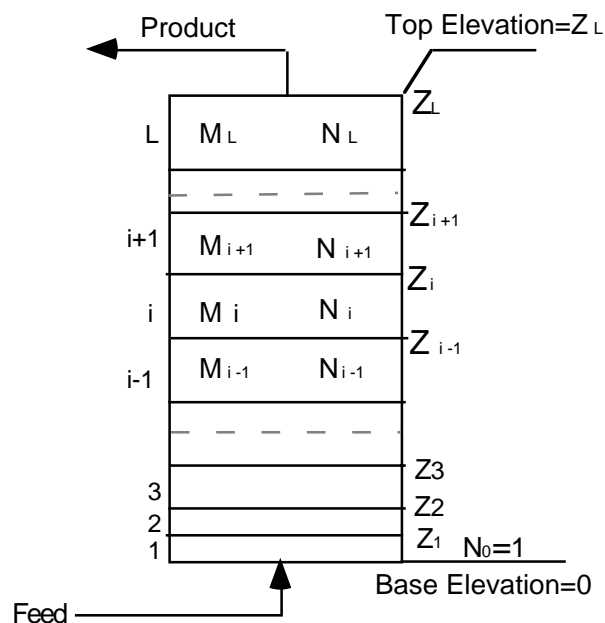
kinetics of any chemical reactions, the time response to varying inputs, time delays, etc.

Fixed Time Zone Methodology (FTZM™) may be applied to eliminate many of the problems inherent in the conventional approach and to systematically design a plug flow reactor model. The fundamental concept in this approach is the application of "time zones" for tracking material through the vessel. A flow vessel is time partitioned into integral multiples, N , of a basic time unit, dt , the integration time step for the simulation. Time zone "i" of size N_i may be defined as a region of the vessel containing a mass or volume of material which has flowed into the vessel over a time period of N_i basic time units. With changes in production rate, feed density and/or composition, the spatial zone boundaries and the number of zones will normally vary with time but the "time" size of each zone is invariant.

FTZM™ Partitioning and Equation Development

Figure 1 illustrates a partitioning of a vessel into L time zones. Depicted in the figure are the zone indices, the elevation at the top boundary of each zone, Z_i , the total mass in each zone, M_i , and the size of each zone, N_i .

Figure 1



Time Zone Partitioning of a Vertical Upflow Vessel

FTZM™ is based on the principle of fluid displacement through the vessel. The fraction of any extensive fluid property such as mass or enthalpy flowing from zone i to zone $i+1$ during a time step is the product of that property and the ratio of dt to the size of zone i ; i.e., $dt/N_i dt = 1/N_i$. Defining this shifting ratio as R_i the total mass flow from zone i to zone $i+1$ is

$$\text{Total Mass Flow} = R_i M_i^{k-1} \quad (1)$$

The total mass and the mass of component j accumulated in zone i at time k are determined, respectively, as

$$M_i^k = (1-R_i)M_i^{k-1} + R_{i-1}M_{i-1}^{k-1} \quad (2)$$

$$M_{ij}^k = (1-R_i)M_{ij}^{k-1} + R_{i-1}M_{i-1j}^{k-1} \quad (3)$$

Equation 1 is the basic transport equation of FTZM™ while equation 2 is a continuity equation. The total enthalpy, H_i^k , of zone i at time k is

$$H_i^k = (1-R_i)H_i^{k-1} + R_{i-1}H_{i-1}^{k-1} \quad (4)$$

For the zonal component mass balances equation 3 is applied for each component j to determine the zone composition. The mass fraction of component j in zone i is computed by

$$mf_{ij}^k = M_{ij}^k / M_i^k \quad (5)$$

From the component masses and component enthalpy functions in conjunction with the total enthalpy from equation 4 the zonal temperature, T_i , may readily be determined. The volume of zone i is

$$V_i^k = \sum_j M_{ij}^k / \rho_{ij}^k \quad (6)$$

where ρ_{ij}^k is the density of component j in zone i. For any zone i in a vertical vessel the elevation, concentration of component j, the mid zone pressure and retention time are computed, respectively, by

$$Z_i^k = Z_{i-1}^k + V_i^k / A \quad (7)$$

$$C_{ij}^k = M_{ij}^k / V_i^k \quad (8)$$

$$P_i^k = P_L^k + g(0.5M_i^k + \sum_{i=i+1}^L M_i^k) / A \quad (9)$$

$$\text{Retention}_{ik} (dt) = \sum_{m=1}^i N_m \quad (10)$$

where A is the cross-sectional vessel area, g is the acceleration of gravity and P_L^k is the pressure at the top of the vessel.

Application of Model Equations

Equation 2 represents the mixing model for a plug flow vessel. The general algorithm for a plug flow reactor requires superposition of reaction kinetic and the associated stoichiometric equations onto these accumulation equations. The foregoing equations may be adapted to various types of plug flow reactors such as combinations of upflow/downflow, pressurized/unpressurized and compressible or incompressible fluid. For all types the shifting equations are sequentially executed from the vessel outlet to feed zones. The product and feed flow rates are determined externally to the vessel. The product mass flow is subtracted from the outlet zone while the feed mass flow is

shifted into the feed zone. A volumetric constraint is imposed on the vessel. If the vessel is filled the total fluid volume and vessel volume must be identical. For a pressurized vessel this continuity constraint is applied to determine the pressure distribution in the vessel. For example, assuming the pressure in a vessel containing gas bubbles is slowly varying the walls of the vessel may be considered rigid and the compressibility of the bubbles will determine the pressure distribution. Therefore with the vessel volume V_v constant, the total volume of solids and liquids, V_s^k and V_L^k , respectively, may be computed by summing the volumes of all solid and liquid components for each zone with equation 6. The total volume of gas by difference is

$$V_g^k = V_v - V_s^k - V_L^k \quad (11)$$

Applying the ideal gas law and neglecting gas solubilities, the gas volume of a vertical upflow vessel as a function of the top pressure is

$$V_{gas}^k = R \sum_i T_i^k / P_i^k \sum_j M_{ij}^k / Mw_j \quad (12)$$

where j denotes gaseous components, R is the universal gas constant, Mw_j is the molecular weight of component j and P_i^k is a function of P_L^k . Newton's method may be applied to adjust P_L^k such that V_{gas}^k is equal to V_g^k .

With all zonal component concentrations and temperatures available, the inherent kinetic and stoichiometric equations are executed and the component masses in each zone updated. In addition the heat released by reaction in each zone, ΔH_{rx} , is added to the total zone enthalpy as computed in equation (3). Thus

$$H_i^k = H_i^{k-1} + \Delta H_{rx}^k \quad (13)$$

All calculations have now been performed for time step k and the sequence is repeated at the next time step, k+1.

FTZM Design of a Plug Flow Reactor Model

Using IDEAS™ a plug flow reactor testing module was developed which enables the model designer to evaluate the dynamics of both the mixing and reaction models. The maximum reaction rate will occur in the feed zone where the reactant concentration(s) are greatest. With knowledge of the maximum expected feed concentrations and temperature the reaction dynamics may be approximated by a first order rate equation

$$dC_i^{k-1} / dt = -K_r C_i^{k-1} \quad (14)$$

where K_r is the rate constant which generates a rate equal to that of the reaction model in the feed zone for the same reactant concentration, C_i^{k-1} . For a constant volumetric feed rate and small reactant concentrations continuity equation

3 may be formulated in terms of reactant concentration and combined with equation 14 to yield

$$C_i^k = C_i^{k-1} + R_i(C_{i-1}^{k-1} - C_i^{k-1}) + \Delta C_i^{k-1} \quad (15)$$

where ΔC_i^{k-1} is the change in reactant concentration due to reaction. Analytically this change is

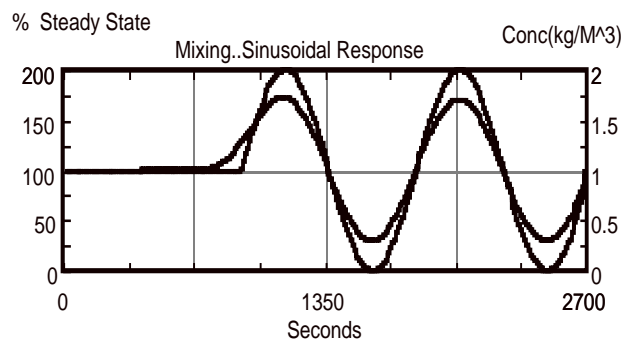
$$\Delta C_i^{k-1} = -C_i^{k-1}(1 - \exp(-K_i dt)) \quad (16)$$

where dt is the basic time step. Equation 15 is the basis for analysis of both mixing and reaction models. For a specified zone size distribution, feed concentration and rate constant, equation 15 is solved sequentially for each zone and the concentration for a specified zone is plotted together with the exact analytical solution for a plug flow reactor at the same retention time.

For mixing model evaluation K_i is set to 0, thus isolating the mixing and reaction models. The model designer may systematically define the size of the time zones through analysis of the mixing model response to pulse, step or sinusoidal changes in the feed concentration. The model dynamics may be evaluated visually by comparison of time delays and response magnitudes with the exact solution or quantitatively using Fourier transform methods. Table 1 is a "zone definition" table illustrating the zone groups with respect to zone number, size and retention. Also displayed is the accumulated retention time up to the end of the group.

Group No.	No. of Zones	Size (dt)	Group Ret.	Total Retention
0	5	51	5	
1	5	2	10	15
2	5	3	15	30
3	10	5	50	80
4	20	10	200	280
5	30	20	600	880
6	40	30	1200	2080

Table 1
Zone Definition Table



Using Table 1 zone definitions Figure 2 demonstrates the mixing model response at the 75th zone to a sinusoidally

Figure 2

Zone Response to Sinusoid in Feed Concentration

varying feed concentration. Both the sinusoid period and ideal retention time are 15 minutes. The attenuation for this frequency is 0.71, the ratio of the amplitude of the response to the exact plug flow solution response. Responses are generally evaluated for total retention times equal to the residence time in the vessel under normal operating conditions. Step responses are valuable for comparing the model and ideal time delay.

The reaction model dynamics are isolated from the mixing dynamics by feeding a constant reactant concentration to the model. The reaction dynamics evaluation consists of comparing the exact solution to that produced by different order numerical integration algorithms in the computation of ΔC_i^{k-1} . For example, the exact solution at a zone may be compared with Euler, 2nd and 4th order Runge Kutta methods. To reduce computational time the lowest order satisfying an accuracy criterion would normally be selected for implementation. The size of the first few zones are normally equal in size to the basic time unit to eliminate mixing effects when the reaction rates are greatest.

Step Responses of a Time Zone

The following derivations reveal the dynamic character of time zones to a step change in volumetric feed rate and to a step change in feed concentration. In the first case the zone volume "adjusts" to accommodate the increase in flow while in the second the zone volume and volumetric feed rates are held constant while the feed concentration changes.

By definition if time zone "i" is at steady state with an inlet flow of Q_0 then the volume of the zone is

$$V_i = Q_0 N_i dt = Q_0 dt / R_i \quad (17)$$

where Q_0 is the volumetric flow into the zone. The zone volume is an extrinsic property of the zone and therefore the transient response of the volume to a step change in Q_0 from Q_0 to mQ_0 at time 0 may be computed from equation 2. At time dt the volume is

$$V_i^1 = mQ_0 dt + V_i^0(1 - R_i) \quad (18)$$

At time kdt the volume is

$$V_i^k = mQ_0 dt \sum_{j=0}^{k-1} (1 - R_i)^j + V_i^0(1 - R_i)^k \quad (19)$$

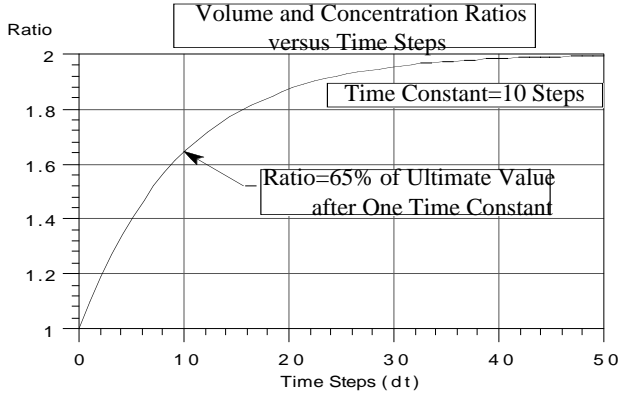
If $R_i = 1$ then the zone attains a volume of $mQ_0 dt = mV_i^0$ in one step. If $R_i < 1$ then $(1 - R_i)^k \rightarrow 0$ in the limit as $k \rightarrow \infty$. Hence as time approaches infinity the second term in equation 19 approaches 0. The first term is the product of a constant and a geometric series with common ratio $1 - R_i < 1$ and therefore a sum equal to $1/R_i = N_i$. Thus in the limit as $k \rightarrow \infty$

$$V_i^\infty = mQ_0 N_i dt = mV_i^0 \quad (20)$$

and the flow out of the zone is

$$Q_{out} = R_i V_i^k = m Q_0 \quad (21)$$

In other words a change in steady state flow into a zone by a ratio "m" results, in the limit, to a new steady state zone volume and outlet flow m times the original values. Figure 4 illustrates a plot of V_i^k/V_i^0 vs time for a step flow change ratio "m" value of 2 and $R_i=0.1$. The response is very



similar to that of a first order system, reaching 65% of its final value after N_i steps; i.e., the zone time constant.

Figure 3
Step Response of a Time Zone

For a step change in feed concentration from C_i^0 to mC_i^0 it can be shown that the zone concentration at time k is defined by an equation very similar to equation 19:

$$C_i^k = m Q_0 dt / V_i^0 \sum_{j=0}^{k-1} (1-R_i)^j + C_i^0 (1-R_i)^k \quad (22)$$

From equation 17 the term $Q_0 dt / V_i^0$ is equal to $1/N_i$, the reciprocal of the zone time constant. Therefore the expression for C_i^k reduces to

$$C_i^k = m C_i^0 / N_i \sum_{j=0}^{k-1} (1-R_i)^j + C_i^0 (1-R_i)^k \quad (23)$$

Using the same reasoning as applied with respect to the volumetric flow step change, in the limit as $k \rightarrow \infty$

$$C_i^\infty = m N_i C_i^0 / N_i = m C_i^0 \quad (24)$$

The associated concentration ratio response curve is identical to the zone volume ratio response curve as illustrated in figure 3, with C_i^k/C_i^0 replacing V_i^k/V_i^0 .

Application of FTZM to an Oxygen Delignification Model

The foregoing methodology was applied to the high fidelity simulation of an oxygen delignification reactor. The feed to the vertical pressurized vessel consists of fibre, caustic and oxygen. The kinetic equations describing the system were presented by L.L. Edwards and M.R. Myers (1). The param-

eters in the kinetic equations (such as activation energies) were determined by the authors from nonlinear, least squares analysis on laboratory and pilot plant data for runs with varying kappa numbers, reaction times, temperatures, pressures, wood species and fractional caustic and oxygen charges on pulp. To best fit the data the lignin was partitioned into three fractions: fast reacting, slow reacting, and bulk/inert. For zone i the simultaneous kinetic equations relating changes in fast and slow kappa numbers, respectively, as a function of temperature, dissolved oxygen, and caustic concentrations are

$$dK_{fi}/dt = -k_{fi} \exp(-E_f/RT_i) [O_2]_i^r [K_{fi}]^s \quad (25)$$

$$dK_{si}/dt = -k_{si} \exp(-E_s/RT_i) [O_2]_i^r [OH]_i^s K_{si} \quad (26)$$

where the total kappa number is

$$K_i = K_{fi} + K_{si} + K_{bi} \quad (27)$$

In these expressions k_{fi} and k_{si} refer to the fast and slow kinetic equation frequency constants, and E_f and E_s , the respective activation energies. The oxygen consumption is linearly related to the amount of lignin reacted by

$$\alpha = \text{mass O}_2 \text{ consumed} / \text{mass lignin reacted} \quad (28)$$

The oxygen solubility was considered to be linearly related to oxygen pressure and exponentially related to caustic concentration (3). The consumption of caustic is not linear (2):

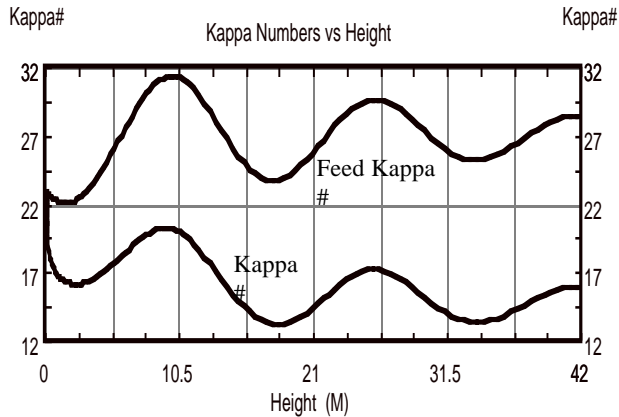
$$\% \text{ NaOH/pulp} = K_{oi} (K_{oi}/K_i - 1) / 19.6 \quad (29)$$

In the caustic consumption expression K_{oi} refers to the total kappa number of the fibre in zone i when the fibre entered the reactor.

The foregoing kinetic model defined the reaction system for the oxygen delignification reactor. After evaluating the mixing model response to changing feed kappa numbers for various zone definitions, equations 2 to 13 were applied along with the foregoing kinetic and stoichiometric equations. Since the O_2 tower was under pressure control the basic time unit for zone definition was chosen as one second.

Figure 4 illustrates the tower profile of a run in which the feed kappa number varied sinusoidally with a period of 30 minutes and an amplitude of 5 units about a mean kappa number of 27. Depicted are the kappa number and the corresponding feed kappa number versus the height from the base of the tower. As expected the kappa no. at any position in the tower is strongly correlated to its kappa number on entering the tower. Because of mixing effects the amplitude attenuation of the inlet kappa number increases with height in the tower. The displacement between

the two curves is due to lignin consumption by reaction, the



difference increasing more rapidly towards the feed inlet where the reaction rate is greatest

Figure 4
Tower and Inlet Kappa Number Profile

Discussion of FTZM™

In the conventional partitioning of a plug flow vessel into fixed volumes, each volume is considered to be a stirred tank about which mass and enthalpy balances are performed. For constant volumetric flow the mixing mass and enthalpy balances for a time zone are identical to those of a stirred tank provided that Euler integration is used for the tank. The size of the time zone in basic time units is identical to the tank time constant.

The output response to a feed concentration change for a model of a series of noninteracting tanks or time zones depends on the order in which the discrete system of equations is solved. In the FTZM™ approach fluid is shifted through the time zones starting at the product zone and finishing at the feed zone. With this computational order a change in zone i as a result of a change in zone $i-1$ is always delayed one time step, thus reflecting a time delay closer to that of plug flow than if the order direction were reversed. In the limit as each tank time constant and therefore each zone size is defined as equal to the basic time step, FTZM™ generates an exact plug flow model, the time delay equaling the number of zones. If the computational order were reversed the time delay would be only one step, independent of the number of zones. However due to computational and memory overhead this zone definition is generally impractical and the plug flow model must be somewhat degraded as a result of increasing zone sizes and thus mixing.

The application of time zones in FTZM™ provides many advantages over the conventional approach. In the conventional method one must be cautious that the size of the feed zone is not smaller than the maximum volumetric feed rate. With FTZM™ this problem is eliminated since the volu-

metric size of the first time zone is *always* equal to the volumetric feed rate. In addition since transport of fluid is based on time and not spatial position, problems in the determination of interzonal flows are eliminated since time zones “adjust” their volumes to accommodate flow changes. Specifically, for a two phase flow of gas and liquid application of the conventional method necessitates iterations on both flows and pressure while FTZM™ iterates only on pressure.

Conclusions

FTZM™ promises to be a valuable tool in the high fidelity modeling of plug flow processes. The modeler may systematically evaluate mixing effects for different time zone definitions as well as quickly evaluate the effect of different order algorithms on the integration of rate equations. As illustrated, under conditions of constant volumetric flow the step response to a concentration change in the flow into a time zone is almost identical to the corresponding step response of a stirred tank or, equivalently, a conventional fixed volume zone. With the dynamic character of time zone volumes many of the difficulties inherent in the fixed volume method are eliminated.

This methodology greatly facilitated the testing and tuning of Simon's Oxygen Delignification Advanced Control package, SODAC™(4). The application of FTZM™ to high fidelity modeling was enhanced by the simulation environment provided in the object oriented platform presented in Part 1, the tool (5).

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