A COMPLETE DYNAMIC MODEL FOR TWIN SCREW EXTRUDERS

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Abstract: A one-dimensional physically motivated dynamic model of a twin-screw extruder for reactive extrusion has been developed. This model can predict extruder behaviour such as pressure, filling ratio, temperature and molar conversion as well profiles as residence time distribution under various operating conditions such as feed rate, screw speed, monomer/initiator ratio and heat flux supplied to the barrel. The model consists of a cascade of perfectly stirred reactors which can be either fully or partially filled with backflow. We consider the mass balance coupled with the momentum balance for the calculation of the pressure profile and the flows between the different reactors. At each reactor is associated the concentration in monomer, the temperature of the matter, the temperatures of the associated piece of screw and barrel. The final model consists of a set of differential algebraic equations. The experimental validation is only made on flow aspects of the model by using experimental RTD's.

Keywords: Reactive extrusion, dynamic model, CSTR, polymerization, residence time distribution, heat transfer

1. INTRODUCTION

A growing interest has emerged in using twin screw extruders as continuous chemical reactors for the polymerisation. Extruders can consequently be used to control the main specific properties of the polymer which are intrinsically linked to the operating pressure and temperature. The primary objective of our research is the control design for polymerization in a twin screw co-rotating extruder. This strategy is applied to the ϵ-caprolactone polymerization. In the case of the regulation of the pressure gradient ΔP at the die, it appears rapidly that the use of simple "grey box" models characterizing an input-output behaviour is not satisfying since the various couplings between all the input variables are not taken into account, see Choulak et al. (2001). This methodology leads to a bunch of simple models, all the bigger as you consider important ranges of variations for manipulated variables (such as feed rate, screw speed, monomer/initiator ratio) since the comportment of the extruder is greatly nonlinear. This approach needs to be able to distinguish between the various models and usually gives rise to synthesize very conservative controls because of the uncertainty of the models. The reader can refer to other publications in the extrusion area

: see Haley and Mulvaney (2000), Tan and Hofer (1995).

Clearly the control of such processes need to have a good understanding of mass flow, rheology, mixing time and thermal behaviour. As far as control is concerned, highly detailed models based on Navier-Stokes equations are not relevant as they are to complicated and do not adress the process modelling but only flowing matter modelling. As far as we know and within the framework of control, he two main references dealing with counter-rotating twin screw extruders are : Ganzeveld et al. (1994) and Graaf et al. (1997) . In these two papers, the description of the material flow along the extruder is intrinsically linked to the model of the C-chamber (which behaves as a CSTR).

In Ganzeveld et al. (1994), the authors consider that the feed in monomer is liquid and then the extruder is formed by two zones : a partially filled zone and a pumping zone (fully filled zone). Thanks to the model of the C-shaped chambers, the pressure and the constant density assumptions, the authors obtain a model issued of mass balance in each chamber represented by differential equations. Moreover the authors couple the energy balance in the barrel to the monomer concentration balance. The main restriction of this model is that there is no accumulation of material in the chambers : the flow profile is fixed. Moreover the length of the fully filled zone is fixed by the die pressure which is an input parameter for the model.

In Graaf et al. (1997), the authors focus on the modelling for predicting the residence time distribution (RTD). The authors consider that the extruder is divided in four zones : the hopper zone (conveying of solid), the partially filled zone (PFZ), a fully filled zone (FFZ) and the die. As previously, the authors obtain a model issued of mass balance in each zone represented by finite differential equations. The main restriction of the model is that the authors fix beforehand the matter occupied volume of each zone; as the result the model cannot predict the flow profile of the extruder and the computation of the gradient pressure is easy.

On the other hand, we find studies concerning food engineering. Four interesting papers emerge : Yacu (1985), Kulshreshta et al. (1991), Kulshreshta and Zaror (1992), and Li (2001). In these four papers are proposed models for twin screw co-rotating extruder taking into account mass and energy balance. The two first papers propose stationary models. The model proposed in Yacu (1985) is the first model predicting axial profile of temperature and pressure in a twin-screw co-rotating extruder. It consists in analytical expressions in the different variables with respect to the spatial coordinate.

It is considered that no heat exchange occurs between the food melt and the screw. Following the zones under consideration (PFZ or CFZ) viscous heat dissipation are negligible or not. In any zone there is heat transfer between the melting and the barrel. The computations of the heat dissipation are based on the work of Martelli (1982). The pressure profile is supposed to be continuous. The computation of the length (related to the pressure profile) of the melt pumping section is made by a trial and error approach. In Kulshreshta et al. (1991), a stationary axial model (from heat and material balances) is presented with the same assumptions. Again the authors consider a simple screw configuration with two zones : the solid conveying zone (PFZ) and the CFZ but their work can be generalized at least theoretically to more than two zones. The continuous treatment of the temperature and pressure profiles leads to a set of differential equations. Again an optimization method is used in order to compute the coordinate of the transition between the two zones in the case of one transition zone. This work was followed by the unsteady version presented in Kulshreshta and Zaror (1992) described by partial differential equations.

In Li (2001) is presented a model for extrusion cooking which is basically the same as in Kulshreshta and Zaror (1992) with two zones (PFZ and CFZ). The difference is in the fact they do not express the mass balance in terms of filling factor but in term of mass flow rate which is continuous. The authors obtain a model described by partial differential equations for temperature and mass flow rate with algebraic constraints for the pressure. The author propose a scheme for integrating these equations.

Finally almost all the authors give models under some restrictive assumptions since many phenomena occur in the extruder :

First, all the authors consider unidirectional analysis. This hypothesis is reasonable since it is not easy to place sensors inside the barrel to measure temperatures, concentrations, flows or information such viscosity. Clearly the model we will build up is also unidirectional. From a process control point of view, the models are often incomplete since energy balance in the screw and the barrel are often not taken into account. It seems to us very important to take these balance into account since in reactive extrusion the problem of controlling the temperature is crucial. We do not found any justification for neglecting their effect.

The second aspect of models is relative to the description of flows along the extruder. The basic constituent of these models is the C-chamber model which is currently used even for co-rotating extruder. This model is the same as a continuously stirred reactor with direct and pressure-back flows. Thus the authors write the mass and energy balance on this C-chamber. From a geometric point of view, this notion corresponds to a channel. Our vision is more global in the sense that we consider a piece of screw rather than a channel. But the computation of flows follows in the same way. On the other hand, simple screw profiles are used in previous quoted papers. The use of these models to more realistic profiles is not an easy task. Our method, more global, permits to take into account more realistic profiles with an acceptable loss of precision, mixing both geometrical and estimated parameters.

In most of the case, precise rheological and kinetic model are not used. In reactive extrusion the hydrodynamic behavior of the melting is strongly influenced by the rheological properties. In general the melt rheology is supposed to be non-Newtonian. But for the computation of flows, Newtonian behavior is supposed. The computations are easier and remain locally valid.

This paper reports a mathematical unidirectional model for twin-screw extruders. The modelling objective is to predict temperature, concentration and pressure profiles at any time and for a large class of operating conditions. Moreover we present the methodology used to achieve our goal. This is based on a decoupled analysis of the hydrodynamic pattern and the geometry of the extruder. In the next section we present the models used to describe the different phenomena occurring in the reactive extrusion process. In section III, we propose a method to obtain the flow model from the RTD and geometric information. The validation of this model is presented from RTD experiments.

2. THE GEOMETRICAL MODEL

2.1 Description of the extruder geometry

The polymerization is carried out in an intermeshing self-wiping co-rotating extruder (Leistritz LSM $30-34$, centreline distance : $C_1 = 30\ mm$, screw diameter : $D = 34\ mm$, barrel length : $L = 1.2\ m$). The extruder barrel is divided into 10 equal zones. Each zone has individual electrical resistance heaters and a water cooling systems. The screw profile is made up of two blocks of kneading discs, direct screw (right handed elements) and one reverse screw (left handed elements) and the die. The dimension of the tubular die are length $= 10\ mm$, and diameter $= 2\ mm$. The pressure sensors allows to determine the pressure gradient inside the die.

2.2 The flow modelling

The backflow reactor model is chosen as the basic element of the model.

This model has the advantage to represent any element (or a part of an element) of the screw (direct screw elements, reverse screw elements, kneading disk block and the die). The kneading disk block can be considered as a direct screw element or a reverse one according to the staggering angle. The three structures of the basic element are presented in figure 1 ; they correspond to the die, the reverse and direct screw configurations.

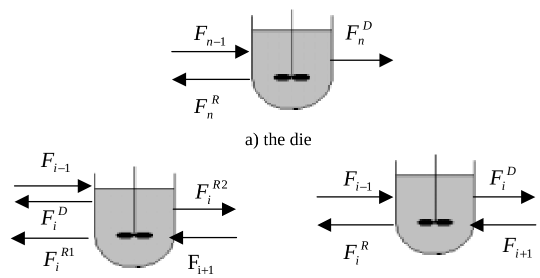

Fig. 1. Structure of the basic elements.

Remark 2.1. The model of the reverse screw element has two pressure depending terms. The F_i^{R1} flow is necessary since without this term the upstream reactors are filled up but not the down stream ones. The flow through the die is considered as being a Poiseuille flow inside a tube.

Let us consider the basic reactor number i as in figure 1.c). This reactor is characterized by its volume and four mass flows. For all these elements, there is no pressure build-up in the partially filled zones and then no reverse flow. In the fully-filled zones, there is a pressure gradient and then a reverse flow occurs. The mass balance for the reactor i leads to :

$$M_i^m \frac{df_i}{dt} = F_{i-1} + F_{i+1} - F_i^D - F_i^R \qquad (1)$$

And for the die :

$$M_n^m \frac{df_n}{dt} = F_{n-1} - F_n^D - F_n^R \qquad (2)$$

The expressions of the direct and reverse flow rates are classical ones obtained from Newtonian hypotheses (see Booy (1980)). The expressions of these flow rates depend on the screw configuration They are given in table 1.

The expressions of the K_i^D's and K_i^R's are functions of the geometry of the screw and of the length of the piece of screw under consideration. The inlet flow rates are equal to the outlet ones when the filling factor of a reactor is equal to one. This leads to the pressure build-up related

Reactor i	F_i^D	F_i^R
Direct pitch	$K_i^D f_i V_i$	if f_i or $f_{i-1} = 1$ $K_i^R(P_i - P_{i-1})$
Reverse pitch	$K_i^D f_i V_i$	if f_i or $f_{i-1} = 1$ $K_i^{R1}(P_{i-1} - P_i)$ if f_i or $f_{i+1} = 1$ $K_i^{R2}(P_i - P_{i+1})$
Die	$K_n^D(P_n - P_a)$	$K_n^R(P_n - P_{n-1})$

Table 1. Expressions of flow rates.(See Booy (1980))

to this reactor and a continuity equation for the mass flow rate. When the filling factor is less than 1, the pressure is supposed to be equal to the atmospheric pressure, say P_a. From a methodological point of view, the computation process is initiated by knowing the profile of filling factor along the extruder. Then the computation of direct flow rate can be done. The profile of pressure is algebraically deduced from the continuity equations (we have to solve a linear system of the type $AP = B$ where A is a matrix, B a vector and P the pressure profile vector. The matrix A is always regular and triangular. When P is obtained, the F_i^R are deduced and equation (1) can be integrated.

2.3 The reaction modelling

The reaction under consideration is the polymerization of the ϵ-caprolactone with tetrapropoxy titanium as initiator. As specified in Gimenez (1999) we consider that the reaction rate is of order 1 with respect to the monomer. The monomer balance is given by equation (3).

$$f_i V_i \mathcal{M} \frac{dC_i}{dt} = (F_{i-1}C_{i-1} + F_{i+1}C_{i+1}) \\ -(F_i^D + F_i^R)C_i - f_i V_i \mathcal{M} K(I_0) e^{-\frac{E}{RT_i^m}} - \mathcal{M} C_i V_i \frac{df_i}{dt} \quad (3)$$

2.4 The thermal modelling

For the sake of simplicity, let us write the energy balance of the melt in the reactor i for direct element (equation (4)). The two first terms of the right member of the equality represent the energy convected by the flowing matter, the third and forth terms, the heat transfer between the melt and the barrel and the heat transfer between the melt and the screw. The two last terms correspond to viscous heat dissipation and the heat flux due to the reaction.

$$\begin{array}{l} f_i M_i^m C_p^m \frac{dT_i^m}{dt} = F_{i-1} C_p^m (T_{i-1}^m - T_i^m) \\ +F_{i+1} C_p^m (T_{i+1}^m - T_i^m) + \alpha_b f_i S_b (T_i^b - T_i^m) \\ +\alpha_s f_i S_s (T_i^s - T_i^m) + f_i \Psi_i + f_i V_i r(-\Delta H) \end{array} \quad (4)$$

In a same way, the energy balance of the associated piece of barrel and of screw can be written.

3. VALIDATION OF THE FLOW MODEL

3.1 The methodology

Let us consider that for a given screw profile, one has discretized the flow by a serial arrangement of N CSTR's with backflows. The model is then able to calculate the time evolutions of the pressure profile, the filling ration profile and the RTD provided that the expressions given in table 1 are sufficiently accurate.

Unfortunately, the number of CSTR's is unknown and expressions given in table 1 are derived from too simple assumptions. Consequently, one has to proceed to some parameters estimation from experimental RTD in order to complete the model. We show that we can predict RTD for new operating conditions after having completed the estimation procedure by using a first set of experimental results. The RTD experiments are performed at steady state.

The screw profile that we use for this study is as follows :

- the matter inlet;	a direct element;
a reverse element;	a direct element;
a kneading block;	a direct element;
the die.	

According to the nature and the position of these elements, one can assume that the kneading block as well as the reverse element are completely filled.

The resistive behavior of these elements and of the die implies that a part of the direct elements are also completely filled. These completely filled parts of the direct elements are situated respectively just before the die, the kneading block and the reverse elements.

A serial of five CSTR's with backflows has proved to be sufficient to represent the completely filled zone of the extruder. This zone corresponds approximatively to the dynamic part of the RTD (The RTD without the delay). The tracer balances corresponding to these five CSTR's are then written as follows :

$$\begin{cases} \rho V \frac{dC_1}{dt} &= FC_{in} + F_2^R C_2 - F_1^D C_1 \\ \rho V_2 \frac{dC_2}{dt} &= F_1^D C_1 - (F_2^D + F_2^R) C_2 + F_3^R C_3 \\ \rho V \frac{dC_3}{dt} &= F_2^D C_2 - (F_3^D + F_3^R) C_3 + F_4^R C_4 \\ \rho V_4 \frac{dC_4}{dt} &= F_3^D C_3 - (F_4^D + F_4^R) C_4 + F_5^R C_5 \\ \rho V \frac{dC_5}{dt} &= F_4^D C_4 - FC_5 - F_5^R C_5 \end{cases}$$

where V_2 is the volume of the kneading block, V_5 the volume of the reverse element and V the volume of the three other CSTR's representing the completely filled zone of the direct elements.

Since RTD's are performed in stationary conditions and all the reactors are CF, we have some equalities between flows F to F_5, cf equation (6).

$$F = F_1^D - F_2^R = \cdots = F_4^D - F_5^R \quad (6)$$

To simplify, we suppose to know the direct flow F_{B2} of the reverse screw zone. Finally we have four unknowns : V, F_2^D, F_3^D, F_4^D. Practically these parameters have been identified.

From the knowledge of the delay in the signal given by the experiment and the feed rate F, the total volume occupied by the matter in the extruder can be computed. From this, the volume occupied by the matter in the PF zones can be deduced.

The partially filled zone of the extruder is also represented by a cascade of CSTR's (it is evident that this partially filled zone is associated to the direct elements). According to relations given in table 1, the reverse flows are equal to 0 in this zone and the tracer balance is as follows for the reactor i :

$$\rho V_{PR} \frac{dC_i}{dt} = FC_{i-1} - FC_i \qquad (7)$$

where V_{PR} is the volume of one CSTR of the partially filled zone (all the CSTR's have the same volume in this zone). The parameter to be estimated is the number of CSTR's necessary to represent the partially filled zone.

One has now to link together the model obtained from the RTD and the geometrical model described by the set of differential algebraic equations. This is done by inserting in the geometrical model at steady state the values of the previously estimated filling ratios. These latter can be computed as soon as a distribution of geometric volume is chosen. It stays a set of equations depending on parameters K_i^D and K_i^R. Fixing the K_i^R's with geometric considerations, and the K_i^D in the CF zone (since these equations are used to compute the pressures), a set of linear equations is obtained. The other K_i^D's are then deduced.

It is clear that this estimation procedure is based on a trial and error method. The discretization based on CSTR's is arbitrarily chosen at the begining and this choice is confirmed by the quality of the results.

3.2 Experimental Validation of the flow modelling

The RTD experiments are carried out with Polypropylene (Polypropylene characteristics are close of the poly-caprolactone characteristics). The estimation procedure is performed by using the experimental RTD obtained for $N = 150$ (rev/mn) and $F = 5$ kg/h (see Figure 2). From that point, we can simulate RTD's obtained under other operation conditions (see Figures 3,4 and 5). The identified model is satisfactory as the time delay is well-fitted as well as the shape of the RTD. The poor fitting of figure 4 can be explained by the fact we choose too big volumes for the CF zone. With more suitable choice, the fitting will be better. One can also see on Figure 2 to 5 a comparison between our fitted model and a so called geometrical model based on the following assumptions :

- the number and volume of the CSTR's are the same as the fitted model;
- the direct and reverse flows are calculated by using equations given in table 1.

It can be seen that this geometrical model is not satisfory due to the lack of precision of the expressions given in table 1. The order of magnitude of the direct and reverse flows is good but one has to correct their theoretical predictions from the fitting results.

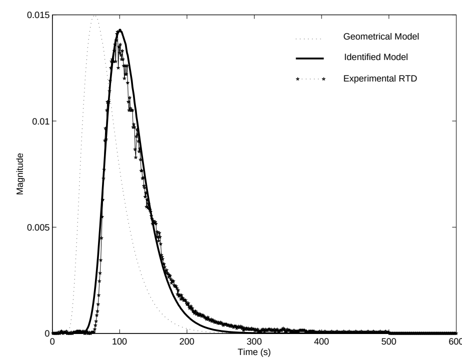

Fig. 2. RTD at screw speed 150 (rev/mn) and flow rate 5 kg/h

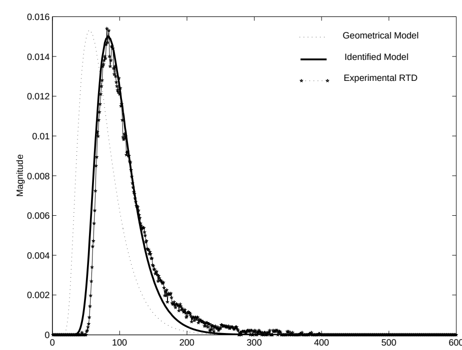

Fig. 3. RTD at screw speed 200 (rev/mn) and flow rate 5 kg/h

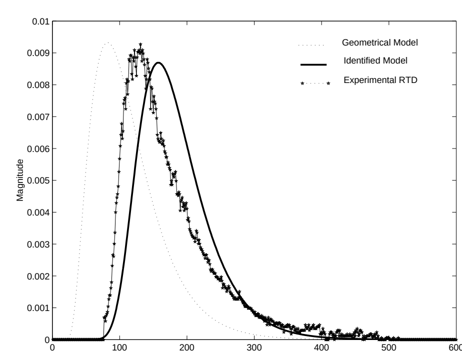

Fig. 4. RTD at screw speed 160 (rev/mn) and flow rate 3 kg/h

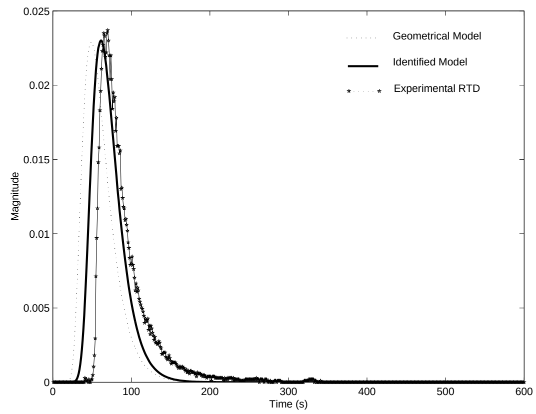

Fig. 5. RTD at screw speed 160 (rev/mn) and flow rate 8 kg/h

4. CONCLUSION

The methodology we have proposed for the extruders dynamic modelling seems to be a right one and give encouraging results. The obtained model can be easily improved by choosing another arrangement of the CSTR's. From the geometric model it can be easily seen that these occupied volumes are different and more important close to the CF zone. Moreover a thermal study of the extruder would give us some additional information on the flow. At this stage, this model can be easily used for control purpose or supervision from a structural point of view even if the model has not been yet entirely validated.

b	: barrel	m	: melt
i	: index of the reactor	s	: screw

Table 2. Subscripts or superscripts.

α_b	: convective heat transfer coefficient of the barrel	$W.m^{-2}.K^{-1}$
α_s	: convective heat transfer coefficient of the screw	$W.m^{-2}.K^{-1}$
Ψ_i	: viscous heat dissipation	W
ΔH	: reaction enthalpy	$J.mol^{-1}$
ρ	: polymer density	$kg.m^3$

Table 3. Greek letters

C_p^m	: specific heat of the melt	$J.kg^{-1}.K^{-1}$
E	: activation energy	$J.mol^{-1}$
f_i	: filling ratio for the reactor i	
F	: feed rate	$kg.s^{-1}$
F_i^D	: direct mass flow rate	$kg.s^{-1}$
F_{i-1}	: mass flow rate coming from the $(i-1)$th reactor	
F_{i+1}	: mass flow rate coming from the $(i+1)$th reactor	
F_i^R	: reverse flow rate produced by reactor i	kgs^{-1}
I_0	: inlet initiator concentration	$mol.m^{-3}$
K	: kinetic constant	s^{-1}
K_i^D	: geometric constant	
K_i^R	: geometric constant	
$\mathcal{M}$	: molar mass of the monomer	$kg.mol^{-1}$
M_i^m	: mass of the melt in the i^{th} reactor	kg
P_a	: atmospheric pressure	Pa
P_i	: i^{th} reactor pressure	Pa
r	: reaction rate	$mol.s^{-1}.m^{-3}$
R	: ideal gas constant	$J.mol^{-1}.K^{-1}$
S_b	: contact surface between the melt and the barrel	m^2
S_s	: contact surface between the melt and the screw	m^2
T_i^m	: temperature of the melt in the reactor i	K
T_i^b	: temperature of the piece of barrel associated to reactor i	K
T_i^s	: temperature of the piece of screw associated to reactor i	K
V_i	: the volume of the reactor i	m^3
N	: rotation speed	$rev.min^{-1}$

Table 4. Notations

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