



# Using splitting methods in continuous digester modeling <sup>☆</sup>

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Received 1 January 2006; received in revised form 1 April 2007; accepted 5 June 2007

## Abstract

The pulp and paper industry plays an important role in European economies. The chemical reactions that transform wood chips in pulp occur mainly in a complex moving bed reactor, the digester. Nowadays the use of mathematical models to simulate the transient behaviour of the digester in terms of temperature and compound concentrations represents a real need for industry because it allows simulation of experiments that can not be afforded or that might be very risky. The digester – the most critical piece of the equipment of a pulp mill – is a heterogeneous reactor with an almost cylindrical shape, where wood chips react with an aqueous solution of sodium hydroxide and sodium sulfide, to remove the lignin from the cellulose fibers. From a mathematical point of view the dynamical behaviour of the reactor can be represented by a system of hyperbolic nonlinear partial differential equations. In this system, with 15 equations, we can identify three main types: the equations that describe the temperature and the concentration respectively of the solid, entrapped liquid and free liquid phase. Each of these type of equations present a certain complexity, its numerical simulation being a hard task. In this sense we point out the high nonlinearity of the functions that represent the chemical reactions; the discontinuities induced by the extraction and injection of the free liquor; the discontinuities in the convection velocity of the free liquor – positive where the liquid flow downwards and negative where the free liquid flows upwards. Numerical methods based on operator splitting, nonuniform refinement and some particular techniques to smooth discontinuities, are studied from a qualitative and quantitative viewpoint. Several simulations on temperature and concentrations of organic and inorganic compounds are presented. Special attention will be devoted to the effects induced in the process by discontinuities of wood chips composition.

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## 1. Introduction

The most critical piece of equipment in a kraft pulp and paper plant is the digester. The digester is a complex heterogeneous reactor where a moving bed of wood chips, containing cellulose, hemicellulose and lignin, reacts with sodium hydroxide and sodium sulfide, in a liquid phase, to remove the lignin from the cellulose

<sup>☆</sup> This work has been supported by Centro de Matemática da Universidade de Coimbra and Project POCTI/35039/MAT/2000.

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fibers. The digester studied in this paper has been described in [1,2] and it is an industrial hydraulic continuous reactor with isothermal cooking, a further development of modified continuous cooking.

Much work has been done for the last decades to model this system, mostly motivated by economical reasons. After the H-factor model of Vroom [3], many other empirical models appeared. The need for a good knowledge of the process promoted the development of a large number of first principles based models, representing different approaches of the reaction kinetics, considering different digester types, or simply making different assumptions. Smith and Williams [4] gave a good contribution including a kinetic model often known as the *Pardue model* and approximated the digester by a series of CSTR (continuous stirred tank reactor). This approach was later followed by other researchers such as Christensen et al. [5] and by Wisniewski et al. [6]. Gustafson et al. [7] proposed a kinetic model in three phases to describe the cooking of a softwood and this work was later extended by Pu [8].

Another interesting work on fundamental modeling of the cooking process of wood is that developed by Foss and co-workers [9–11]. They focused on the second part of two-vessel systems and proposed a mechanistic description based on energy, mass and momentum balances. They used a simplified kinetic model where the initial phase was eliminated and the bulk and residual phases were considered together. As a result of these simplifications the kinetic model used is valid for kappa numbers between 50 and 150. In spite of this and other assumptions in the kinetic model, the digester model created is quite detailed.

The mathematical transient model used here was derived from the fundamental principles of mass and energy conservation and is represented by a system of fifteen nonlinear partial differential equations (PDE) of convection–reaction type. In [2], it is possible to find a transient model of an industrial digester that takes into account many mechanical and processual details inside and outside the digester. In order to study the numerical viability of applying Smolaeskiwicz procedure to this problem, the basic equations of the model developed in [2] were taken. That 15 equations correspond to the mass and energy balances.

The numerical method used in the discretization of this PDE's system is based on operator splitting which essentially consists of considering separately convection and reaction phenomena. Splitting methods have been studied by several authors [12–16]. A mathematical analysis of convergence for the method used in this paper is developed in [17,18].

As far as the specificity of our approach is concerned we point out two main aspects. The system of PDE's presented furnishes a description of the transient behaviour of the digester – important, for example, when grade transitions occur. From a mathematical point of view, the application of splitting methods to the system allow the use of specially tailored methods adapted to the phenomena that take place in each part of the digester.

The paper is organized as follows. In Section 2 we present the mathematical model. In Section 3 we describe the numerical splitting method used in the discretization. Finally in Section 4 numerical simulations are included.

## 2. A mathematical model of the transient behaviour of the digester

The digester studied in this paper (Fig. 1) is a continuous kamyr digester with isothermal cooking. It has an almost cylindrical shape, multiple liquid outputs and intermediate external heating points. The porous wood chips – solid phase – containing cellulose, hemicellulose and lignin react with an aqueous solution – free liquid phase – essentially composed by sodium hydroxide and sodium sulfide which penetrate the wood chips – entrapped liquid phase. The aim of the reaction is the remotion of lignin from the cellulose fibers. In the upper part of the digester the solid and liquid phases flow downwards concurrently. At several levels of the digester's height the free liquid is extracted, enriched and heated before being reinjected at a similar location. This temporary extraction is called circulation. At the main extraction the free liquor is extracted and the wood chips flow then countercurrently with a free liquor introduced at the bottom. This flow washes the degradation products from the pulp and cools it to reduce damage caused by the continuation of the reaction.

The mathematical model under investigation corresponds to the basic equations of the model developed in [2], that is the equations that come from mass and energy balances. It is represented by a set of fifteen PDE's of convection–reaction type involving fifteen dependent variables (concentrations and temperature). The fifteen dependent variables represent the concentrations of organic compounds (cellulose (C), hemicellulose (H) and

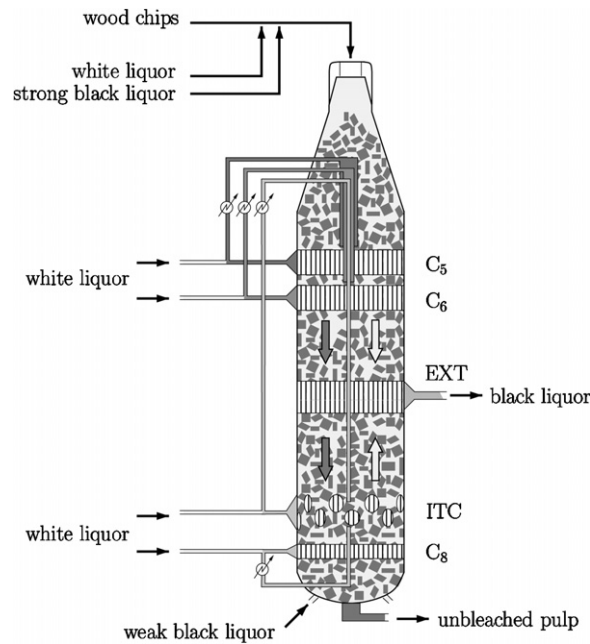


Fig. 1. The digester.

lignin (L)) in each of the three phases and the concentration of the inorganic compounds (effective alkali (E) and hydrogen sulfide ion (S)) in both liquid phases as well as the temperature. Since the wood chips are porous and impregnated with liquor the digester is a reactor with three simultaneous phases (solid, entrapped and free liquors). The notations concerning the variables are described in Table 1.

### 2.1. The equations of the model

We will consider that the digester is a cylindrical plug flow reactor. Consequently, in our model, there is only one space variable,  $z$ . As we may see in the picture of Fig. 1, there are several relevant points along

Table 1  
Notations

Phase	Unknown	Meaning	Units
Solid	$y_1^S$	Mass fraction of C in the chips	dimensionless
	$y_2^S$	Mass fraction of H in the chips	dimensionless
	$y_3^S$	Mass fraction of L in the chips	dimensionless
Entrapped liquid	$y_1^E$	Mass concentration of C in the entrapped liquor	$\text{kg m}^{-3}$
	$y_2^E$	Mass concentration of H in the entrapped liquor	$\text{kg m}^{-3}$
	$y_3^E$	Mass concentration of L in the entrapped liquor	$\text{kg m}^{-3}$
	$y_4^E$	Molar concentration of E in the entrapped liquor	$\text{mol dm}^{-3}$
	$y_5^E$	Molar concentration of S in the entrapped liquor	$\text{mol dm}^{-3}$
	$y_6^E$	Temperature of the entrapped liquor	K
Free liquid	$y_1^F$	Mass concentration of C in the free liquor	$\text{kg m}^{-3}$
	$y_2^F$	Mass concentration of H in the free liquor	$\text{kg m}^{-3}$
	$y_3^F$	Mass concentration of L in the free liquor	$\text{kg m}^{-3}$
	$y_4^F$	Molar concentration of E in the free liquor	$\text{mol dm}^{-3}$
	$y_5^F$	Molar concentration of S in the free liquor	$\text{mol dm}^{-3}$
	$y_6^F$	Temperature of the free liquor	K

the length of the digester. In the case under study we have the following points: the top of the digester, at position  $z_t = 0$  m; the circulation  $C_5$ , at position  $z_{s,C_5} = 8.7$  m, and the injection point just before it, at position  $z_{inj,C_5} = 8.2$  m; the circulation  $C_6$ , at  $z_{s,C_6} = 13.1$  m, and the injection point just before it, at position  $z_{inj,C_6} = 12.5$  m; the main extraction EXT, at position  $z_{s,EXT} = 20.1$  m; the ITC, at position  $z_{s,ITC} = 33.6$  m, and the injection point in it, at the same position  $z_{inj,ITC} = z_{s,ITC}$ ; the circulation  $C_8$ , at position  $z_{s,C_8} = 37.3$  m, and the injection point in it, at the same position  $z_{inj,C_8} = z_{s,C_8}$ , and, finally, the bottom of the digester, at position  $z_b = 41$  m. We will define  $\Omega = \{C_5, C_6, EXT, ITC, C_8\}$ .

### 2.1.1. Solid phase

In order to establish the equations of the model, we start by considering the solid phase. In this phase there is only organic material and, for  $i = 1, 2, 3$ , the concentrations  $y_i^S$  satisfy

$$\frac{\partial y_i^S}{\partial t} = -u_c \frac{\partial y_i^S}{\partial z} + R_i, \quad (1)$$

where  $u_c$  is the interstitial velocity of the moving bed of chips ( $\text{m min}^{-1}$ ) and  $R_i$  the reactions rates ( $\text{min}^{-1}$ ).

The values of the reaction rates,  $R_i$ , have been computed using the kinetic equations proposed by Nóbrega and Castro [19] and they depend not only on the concentrations of the organic material in the solid matrix but also on the concentration of effective alkali in the entrapped phase ( $y_4^E$ ), on the temperature of the chips (solid matrix and entrapped liquor) ( $y_6^E$ ) and on the concentration of hydrogen sulfide ion in the entrapped phase ( $y_5^E$ ). The values of  $R_i$  are defined by:

for cellulose ( $i = 1$ ),

$$R_1 = \begin{cases} -\exp\left(4.16 - \frac{3708.0}{y_6^E}\right) y_4^E y_1^S, & y_3^S \geq 0.185, \\ -\exp\left(27.28 - \frac{14256.9}{y_6^E}\right) y_4^E y_1^S, & 0.02 \leq y_3^S < 0.185, \\ -\exp\left(27.28 - \frac{14256.9}{y_6^E}\right) y_4^E y_1^S, & y_3^S < 0.02; \end{cases} \quad (2)$$

for hemicellulose ( $i = 2$ ),

$$R_2 = \begin{cases} -\exp\left(12.62 - \frac{6454.6}{y_6^E}\right) y_4^E y_2^S, & y_3^S \geq 0.185, \\ -\exp\left(18.75 - \frac{9379.9}{y_6^E}\right) y_4^E y_2^S, & 0.02 \leq y_3^S < 0.185, \\ -\exp\left(5.3 - \frac{7443.4}{y_6^E}\right) y_4^E y_2^S, & y_3^S < 0.02; \end{cases} \quad (3)$$

for lignin ( $i = 3$ ),

$$R_3 = \begin{cases} -\exp\left(6.12 - \frac{4307.7}{y_6^E}\right) y_3^S, & y_3^S \geq 0.185, \\ -\exp\left(35.35 - \frac{16100.0}{y_6^E}\right) y_4^E y_3^S, & \\ -\exp\left(29.23 - \frac{14400.0}{y_6^E}\right) (y_4^E)^{0.03} (y_5^E)^{0.87} y_3^S, & 0.02 \leq y_3^S < 0.185, \\ -\exp\left(19.64 - \frac{9800.0}{y_6^E}\right) (y_4^E)^{1.7} y_3^S, & y_3^S < 0.02; \end{cases} \quad (4)$$

for effective alkali ( $i = 4$ ),

$$R_4 = \frac{1}{R_{lw}} (2R_3 + 16(R_1 + R_2)), \quad (5)$$

where  $R_{lw}$  is a liquid to wood ratio ( $\text{dm}^3 \text{kg}^{-1}$ ), and for hydrogen sulfide ion ( $i = 5$ ),  $R_5 = 0$ .

2.1.2. Entrapped liquid phase

For the entrapped liquid phase, we have, for the concentrations  $y_i^E$ ,  $i = 1, \dots, 5$ , the following equations

$$\epsilon_c \frac{\partial y_i^E}{\partial t} = -u_c \epsilon_c \frac{\partial y_i^E}{\partial z} + \alpha_i R_i + \left( k_{m,i} + u_c \frac{\partial \epsilon_c}{\partial z} \right) (y_i^F - y_i^E), \tag{6}$$

where  $\epsilon_c$  is the porosity of the chips (dimensionless),  $k_{m,i}$  represents the mass transfer of species  $i$  ( $\text{min}^{-1}$ ),  $\alpha_i = -\rho_{c,OD}$ , for  $i = 1, 2, 3$ , and  $\alpha_i = \epsilon_c$ , for  $i = 4, 5$ , where  $\rho_{c,OD}$  is the specific mass of oven dry chips ( $\text{kg m}^{-3}$ ), and the reaction rates  $R_i$  are defined as in the solid phase. The parameter  $\rho_{c,OD}$  has a similar behaviour of that of  $\epsilon_{pi}$ , where  $\epsilon_{pi}$  is the porosity of an outside pile of chips.

According to [1], the term  $\epsilon_c$  is given by  $\epsilon_c = 1 - (y_1^S + y_2^S + y_3^S)(1 - \epsilon_0)$ , where  $\epsilon_0$  is the initial porosity of the chips (dimensionless), which is a function of space and time with the same behaviour of  $\epsilon_{pi}$ . Note that, if we consider different types of wood chips,  $\epsilon_{pi}$  is a piecewise constant function on time which propagates along the digester length with velocity  $u_c$ .

As far as the temperature is concerned, since the heat effects due to chemical reaction are very moderate inside the chips and the contact between solid and entrapped liquid phase is very effective due to the porous structure of the chips, we will assume that the solid matrix and the entrapped liquor are at the same temperature. The temperature behaviour is described by equation

$$\rho_c \tilde{C}_{p,c} \frac{\partial y_6^E}{\partial t} = -u_c \rho_c \tilde{C}_{p,c} \frac{\partial y_6^E}{\partial z} + \left( U^* + \rho_f \tilde{C}_{p,f} u_c \right) (y_6^F - y_6^E) + \rho_{c,OD} (-\Delta H_R) (R_1 + R_2 + R_3). \tag{7}$$

Here  $\rho_c$  (resp.  $\rho_f$ ) is the density of the chips (resp. free liquor), in  $\text{kg m}^{-3}$ ,  $\tilde{C}_{p,c}$  (resp.  $\tilde{C}_{p,f}$ ) is the thermal capacity of the chips (resp. free liquor), in  $\text{kJ m}^{-1} \text{K}^{-1}$ ,  $U^*$  is the heat transfer coefficient, in  $\text{kJ min}^{-1} \text{m}^{-3} \text{K}$ , and  $\Delta H_R$  is the heat of reaction, in  $\text{kJ kg}^{-1}$ .

2.1.3. Free liquid phase

For the free liquid phase, the PDE's that describe the evolution of concentrations have no reaction term because these state variables are simply governed by convection and diffusion between the two liquid phases. For  $i = 1, \dots, 5$ , the free concentrations,  $y_i^F$ , satisfy equations

$$\epsilon_d \frac{\partial y_i^F}{\partial t} = -u_f \epsilon_d \frac{\partial y_i^F}{\partial z} - k_{m,i} (1 - \epsilon_d) (y_i^F - y_i^E) + \beta_{inj} (y_{i,inj}^F - y_i^F), \tag{8}$$

where  $\epsilon_d$  is the porosity of the digester (dimensionless) and  $\beta_{inj} y_{i,inj}^F$  stands for the gains per units of time and of volume of digester due to the injections.

According to [1], the value of  $\epsilon_d$  is given by  $\epsilon_d = 1 - f_c (1 - \epsilon_{pi})$ , where  $f_c$  is, for this digester, experimentally given by

$$f_c = \begin{cases} 0.032z + 1.01, & z < 35.9, \\ 2.16, & z \geq 35.9. \end{cases} \tag{9}$$

The mathematical form of  $\beta_{inj}$  can be found in [2] and is defined as

$$\beta_{inj} = \frac{1}{A_d} \sum_{k \in \Omega - \text{EXT}} \Delta Q_{inj,k} \delta(z - z_{inj,k}), \tag{10}$$

with  $\delta$  the generalized Dirac's delta function. In the previous expression,  $A_d$  represents the sectional area of the digester ( $A_d = 9\pi \text{ m}^2$ ) and  $\Delta Q_{inj,k}$  is the volumetric flow of the injected liquor at the screen  $k$ .

The term  $u_f$ , the interstitial velocity of the free liquor ( $\text{m min}^{-1}$ ), is positive for  $z < z_{s,EXT}$  and negative for  $z > z_{s,EXT}$ .

The value of  $y_{i,inj,k}^F$ , for  $i = 1, \dots, 5$  and  $k \in \Omega - \text{EXT}$ , is given by

$$y_{i,inj,k}^F = y_{i,b}^F + \frac{\Delta Q_{ext,k}}{\Delta Q_{inj,k}} (y_{i,ext,k}^F - y_{i,b,k}^F), \tag{11}$$

where  $y_{i,b,k}$  is the concentration of species  $i$ ,  $i = 1, \dots, 5$ , in the injected white fresh liquor at the screen  $k$  (whose mass flow is known) and

$$y_{i,\text{ext},k}^F = \frac{1}{h_{s,k}} \int_{z_{s,k}}^{z_{s,k}+h_{s,k}} y_i^F dz, \quad (12)$$

where  $h_{s,k}$  is the length of the screen  $k$ . The volumetric flow of the extracted liquor at the screen  $k \in \Omega - \text{EXT}$ ,  $\Delta Q_{\text{ext},k}$ , is the difference between  $\Delta Q_{\text{inj},k}$  and the volumetric flow of the injected white fresh liquor at the same screen (a known value); for  $k = \text{EXT}$ ,  $\Delta Q_{\text{ext},k}$  can be measured directly.

The equation for the temperature is

$$\rho_f \tilde{C}_{p,f} \epsilon_d \frac{\partial}{\partial t} y_6^F = -u_f \rho_f \tilde{C}_{p,f} \epsilon_d \frac{\partial}{\partial z} y_6^F - U^*(1 - \epsilon_d)(y_6^F - y_6^E) + \beta_{\text{inj}} \rho_{\text{inj}} \tilde{C}_{p,\text{inj}} (y_{6,\text{inj}}^F - y_6^F). \quad (13)$$

Note that  $y_{6,\text{inj},k}^F$ , for  $k \in \Omega$ , is a known constant,  $\rho_{\text{inj}}$  ( $\text{kg m}^{-3}$ ) is the specific mass of injected free liquor and  $\tilde{C}_{p,\text{inj}}$  ( $\text{kJ m}^{-1} \text{K}^{-1}$ ) is the thermal capacity of the injected free liquor.

## 2.2. Boundary conditions

As far as the boundary conditions are concerned, we consider  $y_i^S(0, t) = C_i^0(t)$ ,  $i = 1, 2, 3$ , where  $C_i^0(t)$  are known concentrations that can change in time. The concentrations of inorganic compounds and temperature at  $t = 0$  are also assumed to be known. As for  $z \geq z_{s,\text{EXT}}$  the fluid flows upwards and the respective boundary conditions of the dependent variables representing solid and entrapped phases are known in  $z = z_{s,\text{EXT}}$  as a result of the computations in the first part of the digester and the dependent variables concerning the liquid phase are known at  $z = z_b$ .

## 2.3. Initial steady-state

For  $0 \leq z \leq z_{s,\text{EXT}}$  we consider that the digester is filled with chips containing simply water in their pores and consequently, for the organic compounds, we have  $y_i^F(z, 0) = y_i^E(z, 0) = 0$ ,  $i = 1, 2, 3$ , and  $y_i^S(z, 0) = C_i^0(0)$ ,  $i = 1, 2, 3$ , where  $C_i^0(0)$ ,  $i = 1, 2, 3$ , clearly depend on the type of wood the chips are made. For the inorganic compounds, we consider  $y_i^F(z, 0) = y_i^E(z, 0) = 0$ ,  $i = 4, 5$ . For the second part of the reactor, that is  $z_{s,\text{EXT}} \leq z \leq z_b$ , we consider that the digester is filled with a “washing” liquor of known concentration and temperature.

For the temperature, the initial steady-state  $y_6^F(z, 0)$  and  $y_6^E(z, 0)$  are computed using the boundary conditions at  $z = z_t$ . To obtain these steady-state solutions, we will solve the simplified system of ODE's where we consider only the phenomena of mass transfer.

After the first extraction ( $z_{s,\text{EXT}} \leq z < z_b$ ) the steady-state solutions  $y_6^F(z, 0)$  and  $y_6^E(z, 0)$  are computed from the boundary conditions in  $z = z_b$ , for the free liquor, and in  $z = z_{s,\text{EXT}}$ , for the entrapped liquor.

## 3. Numerical method

The PDE's of the digester model can be represented by

$$\frac{\partial y}{\partial t} = F(y), \quad (14)$$

where  $F$  is a nonlinear operator that describes the convection, the reaction and the mass transfer phenomena. In a simplified way, this operator may be consider of type

$$F(y) = -u \frac{\partial y}{\partial z} + f(y), \quad (15)$$

where  $u$  represents a real constant and  $f(y)$  the reaction and mass transfer terms. The initial condition  $y(0, z) = y_0(z)$  is coupled with (14) and (15). In order to take into account the different phenomena (convection and reaction/mass transfer), we will consider a numerical method based on a functional splitting of the problem.

Let us define, in  $[0, T]$ , the splitting grid  $\{t_s\}$  with  $t_s = s\Delta t$  and  $t_{s+1/2} = t_s + \Delta t/2$ , where  $\Delta t$  represents the splitting step size. We suppose that  $y(t)$  is known (at least approximately) at  $t = t_s$ . To compute an approximation for  $y(t)$  at  $t = t_{s+1}$  we decompose (14) into the three subproblems

$$\frac{\partial v}{\partial t} = -u \frac{\partial v}{\partial z}, \quad t \in ]t_s, t_{s+1/2}], \quad v(t_s) = y(t_s), \tag{16}$$

$$\frac{\partial w}{\partial t} = f(w), \quad t \in ]t_s, t_{s+1}], \quad w(t_s) = v(t_{s+1/2}), \tag{17}$$

$$\frac{\partial p}{\partial t} = -u \frac{\partial p}{\partial z}, \quad t \in ]t_{s+1/2}, t_{s+1}], \quad p(t_{s+1/2}) = w(t_{s+1}). \tag{18}$$

It is proved in [18] that, if no boundary condition is considered, that is, if  $z \in \mathbb{R}$ , then  $p(t_{s+1}) = y(t_{s+1})$  which means that there is no functional splitting error. However, if a boundary condition is included at  $z = 0$ , then  $\|p(t_{s+1}) - y(t_{s+1})\|_\infty = O(\Delta t)$ . Under special conditions, this estimation can be improved as mentioned in [14].

### 3.1. Numerical splitting

As problem (16)–(18), for the digester model, exhibits different qualitative behaviour for  $z \leq z_{s,EXT}$  and  $z_{s,EXT} \leq z \leq z_b$ , different families of splitting methods are used in each part of the digester.

For  $z \leq z_{s,EXT}$  the profiles of concentrations and temperature present big gradients due to the sharp changes imposed in the circulations and also to the reaction speeds. To cope with this situation, (16)–(18) is solved with an EIE method obtained by patching together an explicit upwind Smolarkiewicz method – E – to solve (16) and (18) and the implicit Euler method – I – to solve (17). The resulting EIE splitting method is defined by

$$\frac{v_j^{s+1/2} - v_j^s}{\Delta t/2} = -u \left( 1 + \left( 1 - \frac{u\Delta t}{2h} \right) \frac{v_j^s - v_{j-1}^s}{\epsilon + v_j^s + v_{j-1}^s} \right) \frac{v_j^s - v_{j-1}^s}{h}, \quad v_j^s = y_j^s, \tag{19}$$

$$\frac{w_j^{s+1} - w_j^s}{\Delta t} = f(w_j^{s+1}), \quad w_j^s = v_j^{s+1/2}, \tag{20}$$

$$\frac{p_j^{s+1} - p_j^{s+1/2}}{\Delta t/2} = -u \left( 1 + \left( 1 - \frac{u\Delta t}{2h} \right) \frac{p_j^{s+1/2} - p_{j-1}^{s+1/2}}{\epsilon + p_j^{s+1/2} + p_{j-1}^{s+1/2}} \right) \frac{p_j^{s+1/2} - p_{j-1}^{s+1/2}}{h}, \quad p_j^{s+1/2} = w_j^{s+1}, \tag{21}$$

where we assume that  $u > 0$  and  $\epsilon$  is a small positive constant.

For  $z \geq z_{s,EXT}$  the profiles are softer because practically no chemical reaction occurs and consequently a EEE method obtained by patching together three explicit methods can be used. Eqs. (16) and (18) are discretized with the Smolarkiewicz upwind method but where now, as the convection speed  $u < 0$ , we consider

$$\frac{\partial v}{\partial z}(z_j, t_s) \simeq \frac{v_{j+1}^s - v_j^s}{h}, \quad \frac{\partial p}{\partial z}(z_j, t_{s+1/2}) \simeq \frac{p_{j+1}^{s+1/2} - p_j^{s+1/2}}{h}.$$

The reaction Eq. (17) is solved with the explicit Euler method. The global EEE method is then defined by

$$\frac{v_j^{s+1/2} - v_j^s}{\Delta t/2} = -u \left( 1 + \left( 1 - \frac{u\Delta t}{2h} \right) \frac{v_{j+1}^s - v_j^s}{\epsilon + v_j^s + v_{j+1}^s} \right) \frac{v_{j+1}^s - v_j^s}{h}, \quad v_j^s = y_j^s, \tag{22}$$

$$\frac{w_j^{s+1} - w_j^s}{\Delta t} = f(w_j^s), \quad w_j^s = v_j^{s+1/2}, \tag{23}$$

$$\frac{p_j^{s+1} - p_j^{s+1/2}}{\Delta t/2} = -u \left( 1 + \left( 1 - \frac{u\Delta t}{2h} \right) \frac{p_{j+1}^{s+1/2} - p_j^{s+1/2}}{\epsilon + p_j^{s+1/2} + p_{j+1}^{s+1/2}} \right) \frac{p_{j+1}^{s+1/2} - p_j^{s+1/2}}{h}, \quad p_j^{s+1/2} = w_j^{s+1}. \tag{24}$$

### 3.2. Qualitative behaviour

Methods EIE and EEE are globally first order methods. The qualitative behaviour of EEE and EIE is studied in [17].

One of the main advantages of splitting techniques is the fact that all combinations of methods with different explicitness/implicitness and order properties are allowed. In the numerical simulation of the digester behaviour the two first order approaches – EIE and EEE – lead to enough accuracy. In cases where the model exhibits significant stiffness, different choices are more suitable as, for example, second order implicit methods for reaction Eq. (17) and nonlinear flux limiters methods for (16) and (18).

The proposed numerical scheme is a splitting method which treats differently the convection terms and the reaction/mass transfer terms. It is well known (20) that the classical linear schemes, central or upwind, present some drawbacks in respect their qualitative behaviour. For instance, first order upwind can be very diffusive and central difference schemes may exhibit an oscillatory behaviour.

In order to minimize these effects, we will consider the nonlinear Smolarkiewicz upwind scheme to treat the convection part. This method uses the first order Euler-upwind as a basis and introduces a correction step in order to minimize the diffusivity behaviour. More specifically, if we consider the modified equation for the first order Euler-upwind scheme applied to the convection Eq. (16), we conclude that this method represents a second order approximation of the modified equation

$$\frac{\partial y}{\partial t} = -u \frac{\partial y}{\partial z} + \frac{uh}{2} \left( 1 - \frac{u\Delta t}{2h} \right) \frac{\partial^2 y}{\partial z^2}.$$

The term

$$\frac{uh}{2} \left( 1 - \frac{u\Delta t}{2h} \right) \frac{\partial^2 y}{\partial z^2} \quad (25)$$

is responsible for the large diffusivity of the method. In order to remove this diffusive behaviour, we will consider (25) as a convective term of the form  $-\frac{\partial u^* y}{\partial z}$ , with

$$u^* = -\frac{uh}{2y} \left( 1 - \frac{u\Delta t}{2h} \right) \frac{\partial y}{\partial z}. \quad (26)$$

The idea of Smolarkiewicz [20] was to add another upwind step using the additional velocity  $u^*$  with minus sign. Doing this, we obtain a second order scheme to discretize (16) which is less diffusive than the Euler-upwind scheme. The anti-diffusive velocity  $u^*$  is approximated numerically by

$$u_j^* \approx u \left( 1 - \frac{u\Delta t}{2h} \right) \frac{y_j - y_{j-1}}{y_j + y_{j-1}}.$$

In order to study the numerical stability of the proposed numerical methods, let us consider  $\tilde{v}_h^s$ ,  $\tilde{w}_h^s$  and  $\tilde{p}_h^s$  to be perturbed numerical solutions of (19)–(21), respectively. Let

$$J_f = \left[ \frac{\partial f^{(i)}}{\partial y_j} (\theta_j^{s+1}) \right],$$

with

$$\theta_j^{s+1} = \left( \tilde{w}_1^{s+1}, \dots, \tilde{w}_{j-1}^{s+1}, \theta \tilde{w}_j^{s+1} + (1 - \theta) \tilde{w}_j^{s+1}, \tilde{w}_{j+1}^{s+1}, \dots, \tilde{w}_{n-1}^{s+1} \right), \quad \theta \in (0, 1).$$

With  $y_h^{s+1} = p_h^{s+1}$  and  $\tilde{y}_h^{s+1} = \tilde{p}_h^{s+1}$  Proposition 1 can be proved.

**Proposition 1.** Let  $y_h^{s+1}$  and  $\tilde{y}_h^{s+1}$  be two numerical approximations of  $y(t_{s+1})$  computed from the approximations  $y_h^s, \tilde{y}_h^s \in B_\rho(y(t_s))$ , where  $B_\rho(y(t_s))$  is the open ball with center  $y(t_s)$  and radius  $\rho \in \mathbb{R}^+$ , using the EIE method (19)–(21). Then

$$\|y_h^{s+1} - \tilde{y}_h^{s+1}\|_\infty \leq \|(I - \Delta t J_f)^{-1}\|_\infty^2 (r + r|1 - r|M + |1 - r - r|1 - r|M|) \|y_h^s - \tilde{y}_h^s\|_\infty$$

where  $M = \|\nabla g\|_\infty$ , with  $g(x_1, x_2) = \frac{(x_1 - x_2)^2}{x_1 + x_2}$  and  $r = \frac{u\Delta t}{2h}$ .



According to the previous result, we may conclude that, the EIE method is stable if

$$r \leq \min \{1, 1/M, 1/\|J_f\|_\infty\}. \quad (27)$$

Taking into account that the numerical method (19)–(21) is consistent with the problem (16)–(18), we may conclude that, if  $r$  verifies (27), the numerical solution obtained by EIE, when  $h$  and  $\Delta t$  goes to zero, converges to the exact solution of (16)–(18). But, as we had already mentioned,  $\|p(t_{s+1}) - y(t_{s+1})\|_\infty = O(\Delta t)$ , where  $p$

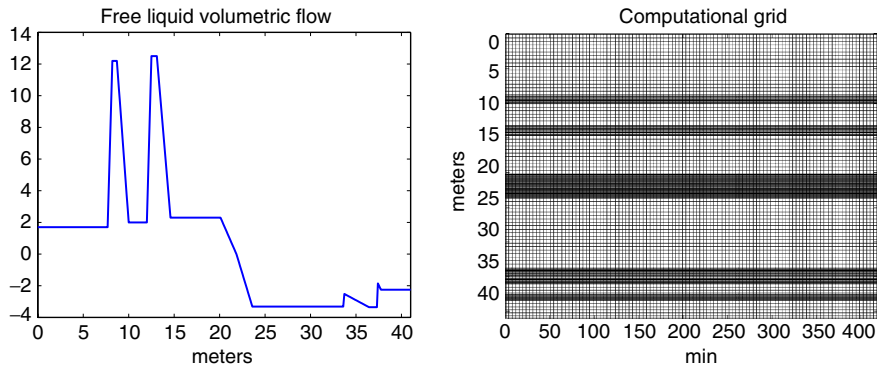


Fig. 2. Free liquid volumetric flow and used grid.

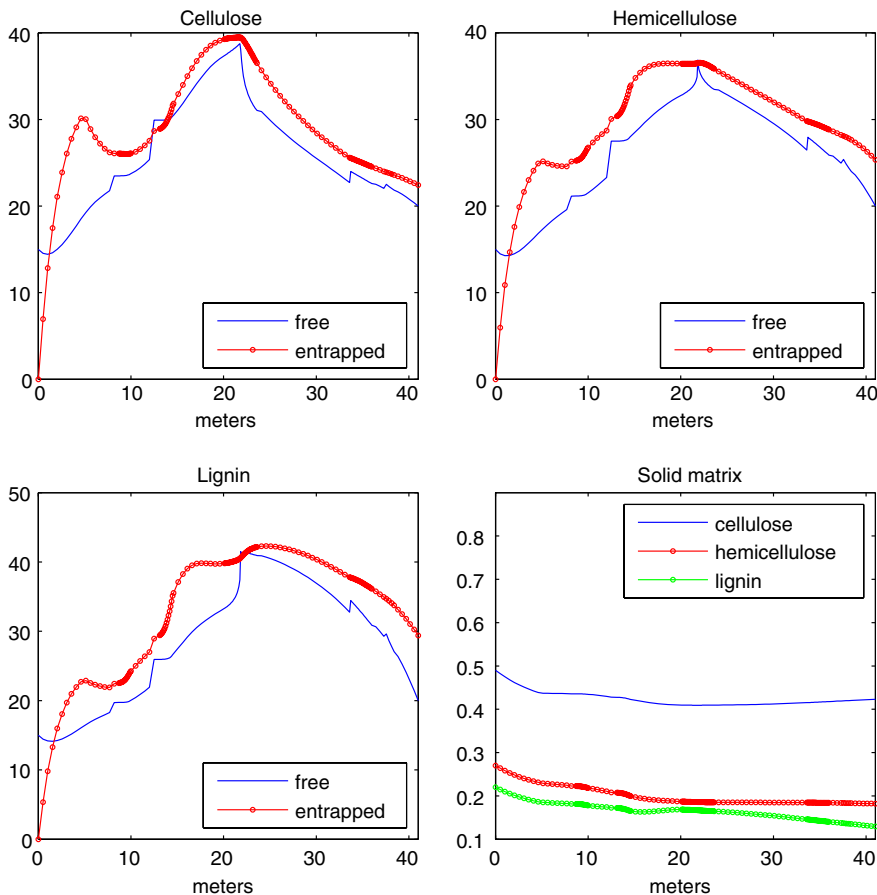


Fig. 3. Inorganic compounds.

and  $y$  are, respectively, the solutions of (16)–(18) and (14)–(15). Then the convergence of the numerical solution obtained by EIE to the solution of the original problem (14)–(15) is concluded.

For the EEE numerical method (22)–(24), an analogous result can be established.

In order to compute an approximation to  $y(x_j)$  when  $y_j = y_{j+1} = 0$ , we replace (26) by

$$u_j^* \approx u \left( 1 - \frac{u\Delta t}{2h} \right) \frac{y_j - y_{j-1}}{\epsilon + y_j + y_{j-1}},$$

where  $\epsilon$  is a small number (for instance  $10^{-15}$ ).

### 3.3. Numerical results

In this section we present some numerical results obtained by the algorithm described in the previous section when applied to the solution of the fifteen PDE's that constitute the core of the digester model. The initial profiles and boundary conditions are defined according to the explanation given in Section 2.

In order to define the computational grid, we took into account the stability result (27). Note that (27) imposes a restriction to the choice of the step sizes which depends on the convection term and on the stiffness of the problem. In our case, we took into account the changes on the free liquid volumetric flow expressed in Fig. 2. In the regions where the flow presents sharp gradients, we refine the spatial grid. Doing this we may consider a constant time step in order to guarantee the stability.

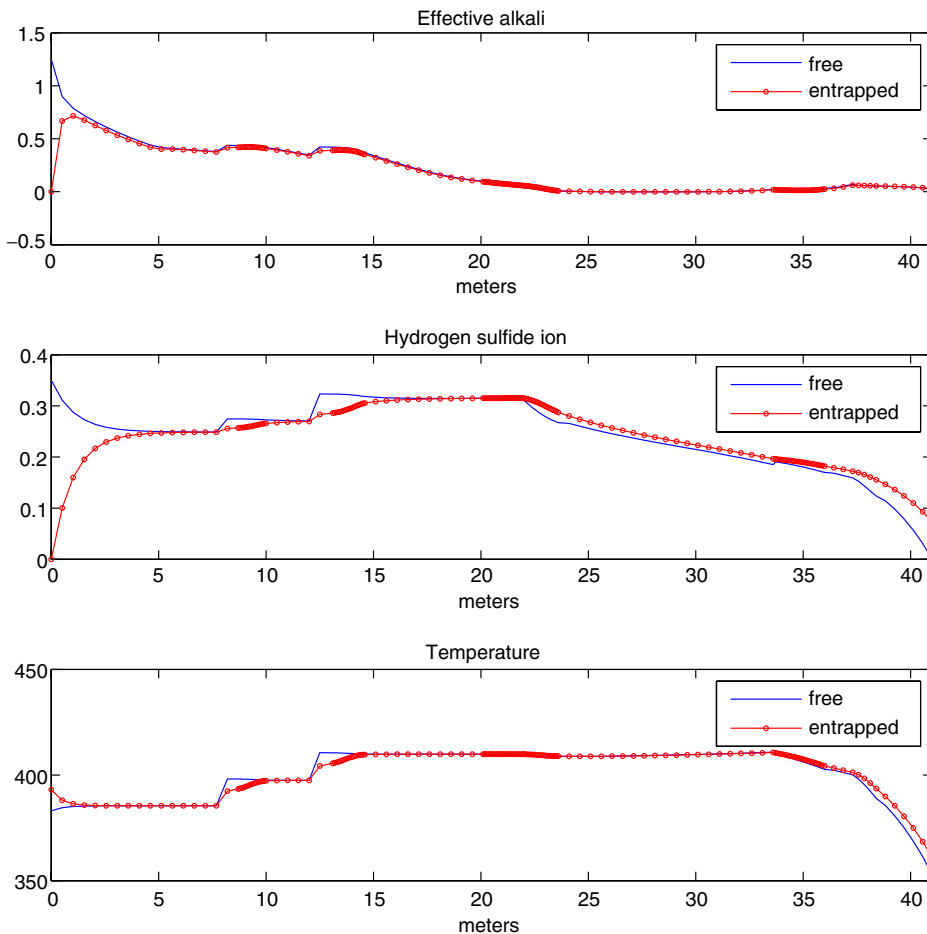


Fig. 4. Inorganic compounds.

To validate this model and the numerical procedure, we did not use real data. Apart from being a model focusing only in the main equations, dynamical data of the digester is not easy to obtain. Meanwhile, in [1] the authors presented a model for the steady-state regime and they used real data. In [21] we presented some numerical results for the steady-state solutions which show physical evidence and agree with the numerical results in [1]. These results are now obtained by the proposed algorithm and are plotted in Figs. 3 and 4.

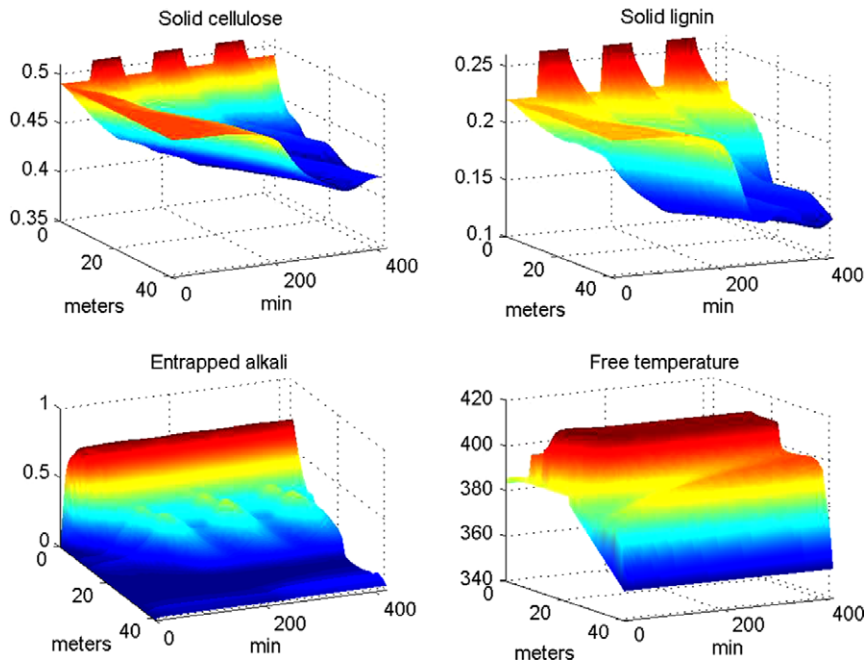


Fig. 5. Changes every hour.

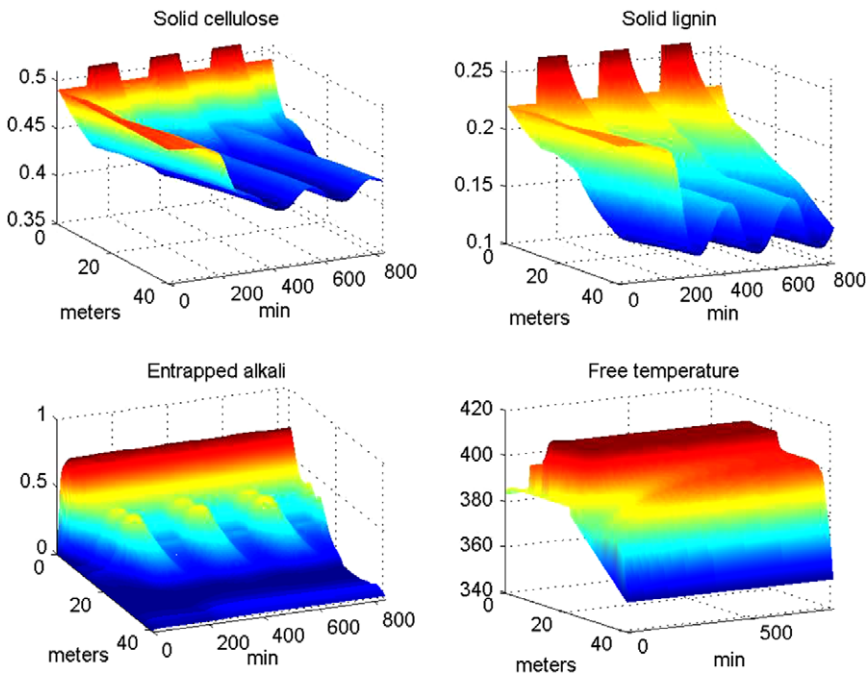


Fig. 6. Changes every two hours.

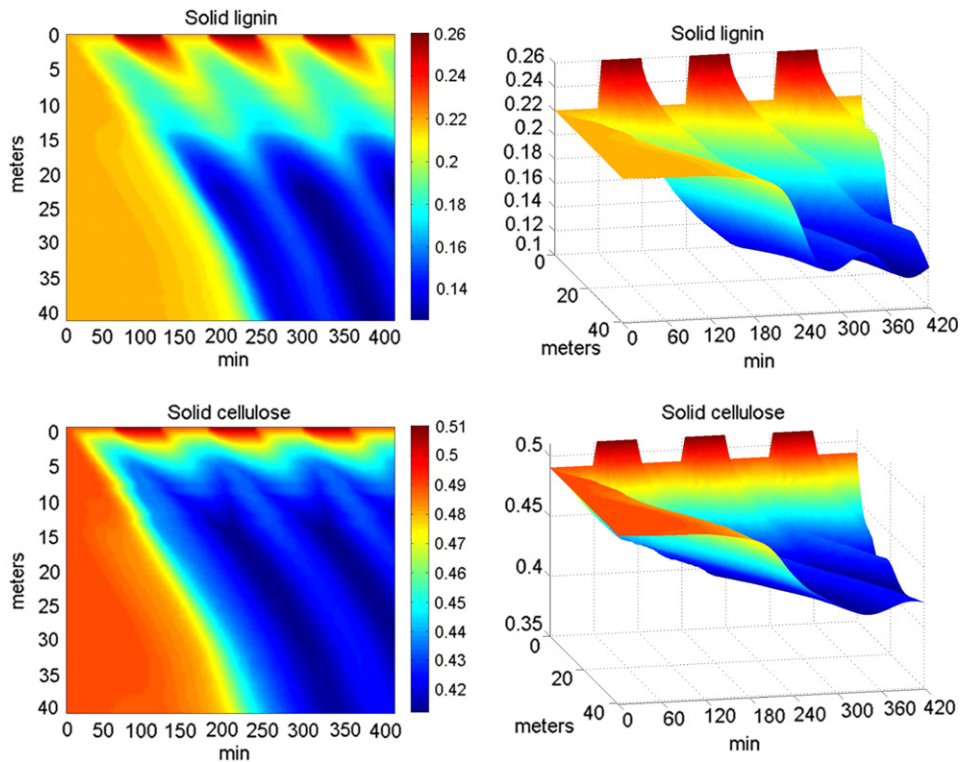


Fig. 7. Changes every hour.

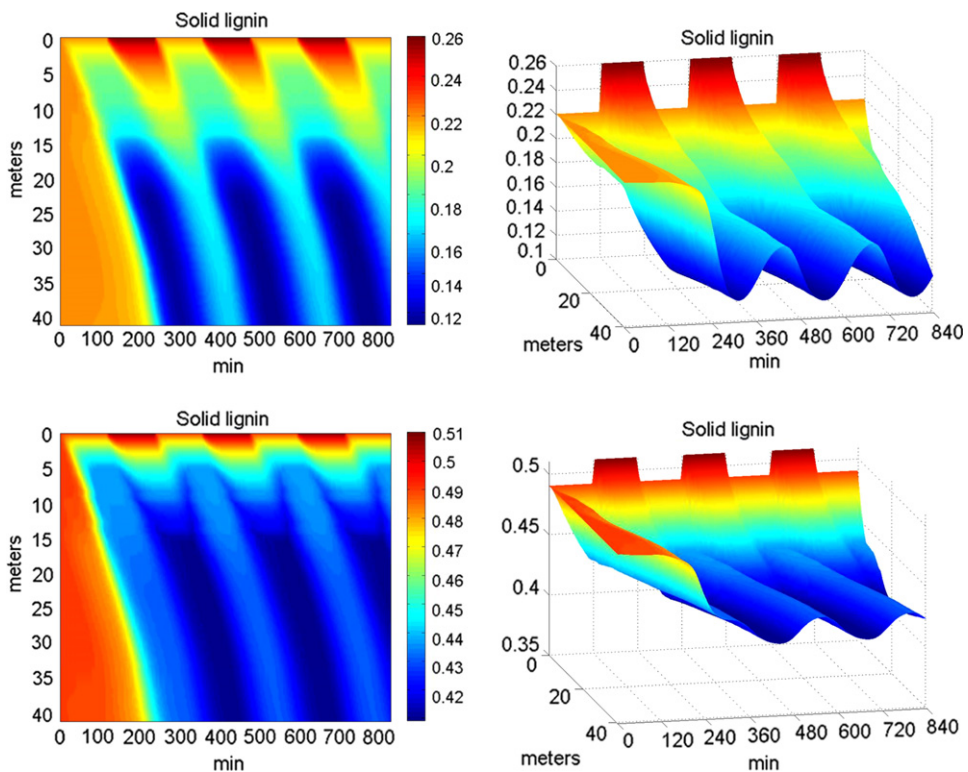


Fig. 8. Changes every two hours.

In the present work our goal is to study the transient behaviour in the case where two different types of wood chips are considered. The evolution in time of the concentrations and temperature are plotted in Figs. 5 and 6, when we change the wood every hour and every two hours. Note that, in spite of the changes been made only for the organic compounds, the inorganic compounds are also affected (see the results for the entrapped alkali). Meanwhile, the temperature inside the digester is not affected by that changes.

In Figs. 7 and 8 we compare the results in solid cellulose and solid lignin when we change the wood chips every hour and every two hours. The periodicity chosen has no correspondence to an industrial context. Our goal is to put in evidence the behaviour of the transient regime. As we may see, for this case, the digester achieves a steady-state regime in less than two hours. This fact is consistent with other computations that we considered. So we may conclude that, the process tends quickly to a steady-state.

We may conclude that, in agreement with physical evidence, the quality of the pulp is affected by the wood changes and the model described here can be used to predict the mean composition of the produced pulp.

## Appendix

The parameters are given in the following table

Parameter	Meaning	Units
$A_d$	Sectional area of the digester	$m^2$
$\epsilon_0$	Initial voidage of the chips	dimensionless
$\epsilon_{pi}$	Voidage of an outside pile of chips	dimensionless
$\epsilon_d$	Voidage of the bed	dimensionless
$\epsilon_c$	Voidage of the chips	dimensionless
$u_c$	Interstitial velocity of the moving bed of chips	$m \text{ min}^{-1}$
$u_f$	Interstitial velocity of the free liquor	$m \text{ min}^{-1}$
$\rho_{c,OD}$	Density of an oven dry chip	$kg \text{ m}^{-3}$
$\rho_c$	Density of the chips	$kg \text{ m}^{-3}$
$\rho_f$	Density of the free liquor	$kg \text{ m}^{-3}$
$\tilde{C}_{p,c}$	Thermal capacity of the chips	$kJ \text{ m}^{-1} \text{ K}^{-1}$
$\tilde{C}_{p,f}$	Thermal capacity of the free liquor	$kJ \text{ m}^{-1} \text{ K}^{-1}$
$k_{m,i}$	Mass transfer coefficient of species $i$	$\text{min}^{-1}$
$U^*$	Heat transfer coefficient	$kJ \text{ min}^{-1} \text{ m}^{-3} \text{ K}$
$R_{lw}$	Liquid to wood ratio	$\text{dm}^{-3} \text{ kg}^{-1}$
$h_{s,k}$	Length of the screen $k$	$m$
$\rho_{inj}$	Density of the injected liquor	$kg \text{ m}^{-3}$
$C_{p,inj}$	Thermal capacity of the injected liquor	$kJ \text{ m}^{-1} \text{ K}^{-1}$
$\Delta Q_{ext,k}$	Volumetric flow of the extracted liquor at screen $k$	$m^3 \text{ min}^{-1}$
$\Delta Q_{inj,k}$	Volumetric flow of the injected liquor at screen $k$	$m^3 \text{ min}^{-1}$

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