UNIVERSIDADE DE VIGO

DEPARTAMENTO DE MATEMÁTICA APLICADA II

OPERATIONAL MODELS FOR REAL TIME APPLICATIONS OF NONLINEAR AND MOVING BOUNDARY PROCESS DISTRIBUTED SYSTEMS

Modelos operacionais para aplicacións en tempo real de sistemas de proceso distribuídos nonliñais e de fronteira móbil

Memoria relizada no Instituto de Investigacións Mariñas (IIM-CSIC) baixo a dirección do Profesor de Investigación Dr. D. Antonio Álvarez Alonso para optar ó grao de Doutor con Mención Internacional pola Universidade de Vigo

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Motivation and Objectives

Thermal processes are widely employed in both Food and Biotechnology industries. Nevertheless, their operation and decision making is still ruled by rigid procedures which are difficult to adapt to sudden changes in the production conditions or to disturbances in the process caused by unforeseen events.

Besides, it must be considered too, that in most of the cases, those operation policies have been not designed according to optimality principles, and therefore there exist opportunities to improve not only the process costs but also the final product quality.

It is in this framework where the development and implementation of real-time oriented mathematical models, which would be employed as the core of computational tools and methods for optimal on-line control processes, constitutes an invaluable mechanism.

Many are the advantages related to process modelling: from providing a virtual environment where new procedures and equipment can be evaluated to predicting the effects of failures or malfunctioning conditions on quality product. However the specific requirements of real-time tasks call for low-dimensional models aimed at reducing the computational times associated to its numerical solving.

In response to this need, the concept of operational model emerges in the sense of a simplified while accurate representation of the system, based on first-principles, whose structure and dimensionality enable its role as the body of control and optimisation on-line schemes which would supply with the desired flexibility to the operation process.

Thus, the objectives of this dissertation will be, in one hand, to provide with the
insights of the methodology proposed, and in the other, to show the applicability and advantages of this integral modelling approach by means of examples consisting of real-time applications for different processes of interest in Food and/or Biotechnology industries.
### Abbreviations

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<td>ALE</td>
<td>Arbitrary Lagrangian-Eulerian</td>
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<td>CVP</td>
<td>Control Vector Parametrisation</td>
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<td>DO</td>
<td>Dynamic Optimisation</td>
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<td>DPS</td>
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<td>FDM</td>
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<td>FTCS</td>
<td>Forward Time Centered Space</td>
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<td>LHS</td>
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<td>ROM</td>
<td>Reduced Order Method</td>
</tr>
</tbody>
</table>
Greek letters

- $\alpha$: Thermal diffusivity
- $\beta$: Control discretisation in CVP
- $\gamma$: Moving front associated function
- $\delta$: Variation in time or space
- $\varepsilon$: Void volume fraction
- $\zeta$: Spatial coordinates
- $\theta$: Auxiliary time variable
- $\lambda$: Eigenvalue
- $\mu$: Viscosity
- $\xi$: Dimensionless independent spatial variable
- $\rho$: Density
- $\sigma$: Stefan-Boltzmann constant
- $\tau$: Dimensionless independent temporal variable
- $\phi$: Global basis function
- $\varphi$: Local basis function
- $\psi$: Test function
- $\Gamma$: Boundary of a spatial domain
- $\Lambda$: Diagonal matrix of eigenvalues
- $\Omega$: Spatial domain with boundary $\Gamma$
- $\Theta$: Dimensionless time scale

Mathematical Operators

- $\Delta(\cdot)$: Laplacian spatial operator $\frac{\partial^2}{\partial \zeta_1^2} + \frac{\partial^2}{\partial \zeta_2^2} + \frac{\partial^2}{\partial \zeta_3^2}$
- $\nabla(\cdot)$: Nabla spatial operator $\frac{\partial}{\partial \zeta_1} + \frac{\partial}{\partial \zeta_2} + \frac{\partial}{\partial \zeta_3}$
- $\langle f, g \rangle_\Omega$: Inner product $\int_\Omega f(\zeta)g(\zeta)d\zeta$
- $||f||_\Omega$: Norm $\langle f, f \rangle_\Omega^{1/2}$
### Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \dot{a} )</td>
<td>Time derivative of variable ( a )</td>
</tr>
<tr>
<td>( \hat{a} )</td>
<td>Dimensionless variable ( a )</td>
</tr>
<tr>
<td>( \tilde{a} )</td>
<td>Approximated variable ( a )</td>
</tr>
<tr>
<td>( f, g, h )</td>
<td>Generic functions</td>
</tr>
<tr>
<td>( i, j, k )</td>
<td>Indexes</td>
</tr>
<tr>
<td>( l )</td>
<td>Number of snapshots</td>
</tr>
<tr>
<td>( \vec{n} )</td>
<td>Normal vector</td>
</tr>
<tr>
<td>( N )</td>
<td>Nodes in the spatial mesh</td>
</tr>
<tr>
<td>( t )</td>
<td>Independent time variable</td>
</tr>
<tr>
<td>( t_{ini} )</td>
<td>Initial time</td>
</tr>
<tr>
<td>( t_f )</td>
<td>Final time</td>
</tr>
</tbody>
</table>
Partial Differential Equations (PDE)

A Partial Differential Equation (PDE) is an equation involving one or more unknown functions of the independent variables together with their partial derivatives. Generically, it can be described as follows:

\[ f \left( \zeta_1, \zeta_2, \zeta_3, u, t, \frac{\partial u}{\partial t}, \frac{\partial^2 u}{\partial t^2}, \frac{\partial u}{\partial \zeta_1}, \frac{\partial^2 u}{\partial \zeta_1 \partial \zeta_2}, \ldots, \frac{\partial u}{\partial \zeta_3}, \frac{\partial^2 u}{\partial \zeta_1 \partial \zeta_3}, \ldots, \right) = 0 \]

where \( \vec{\zeta} = (\zeta_1, \zeta_2, \zeta_3) \in \mathbb{R}^3 \) and the distributed field noted by \( u(\vec{\zeta}, t) \in \mathbb{R} \) is defined over the time interval \( t = [0, +\infty) \).

Usually, PDE’s are classified according to its order. Typically, most of the relevant equations in physics and engineering involve first and/or second order partial derivatives. For the particular case of a second order PDE in \( \mathbb{R}^3 \) of the form:

\[ \sum_{i,j=1}^{3} a_{i,j} \frac{\partial^2 u}{\partial \zeta_i \partial \zeta_j} + \sum_{i=1}^{3} b_i \frac{\partial u}{\partial \zeta_i} + cu = 0 \]  \( \text{(1)} \)

an additional classification can be established attending to the eigenvalues of the matrix \( A = (a_{i,j}) \). Noting by \( p \) the positive eigenvalues and by \( s \) the negative ones:

- **Elliptic**: if \((p, s) = (3, 0)\) or \((p, s) = (0, 3)\). Laplace equation can be found within this group. It must be mentioned that this kind of equations does not involve time derivatives in its formulation.

- **Hyperbolic**: if \( p + s = 3 \), being \( p, s > 0 \). The Wave equation is included into
this type.

• **Parabolic**: if \( p + s < 3 \). As example, the Fourier equation can be mentioned.

Associated to the PDE governing equations, boundary as well as initial conditions must be imposed in order to ensure the existence of a solution. Regarding the boundary conditions, three are the existing types:

• **Dirichlet**: \( u = g \) in \( \Gamma, \forall t \).
• **Neumann**: \( \overrightarrow{n} \nabla u = g \) in \( \Gamma, \forall t \).
• **Robin or mixed**: \( hu + \overrightarrow{n} \nabla u = g \) in \( \Gamma, \forall t \).

where \( g, h \) are generic functions and \( \overrightarrow{n} \) represents the outward normal unit vector with respect to the boundary \( \Gamma \).

On the other hand, initial conditions can be defined for the field and/or its time derivative when required.

**Norm and Normed vector space**

Let \( V \) be a real vector space. A function \( \| \cdot \| : V \to \mathbb{R}^+ \) is a norm on \( V \) if it satisfies:

1. \( |v| \geq 0, \forall v \in V; \)
2. \( |v| = 0 \iff v = 0, \forall v \in V; \)
3. \( |v + w| \leq |v| + |w|, \forall v, w \in V; \)
4. \( |av| = |a||v|, \forall a \in \mathbb{R}, v \in V; \)

A vector space \( V \) together with a norm \( \| \cdot \| \) it is called normed vector space.

**Inner product and Inner Product Space**

A function \( \langle \cdot \rangle : V \to \mathbb{R} \) is an inner product if it satisfies:

1. \( \langle v + w, r \rangle = \langle v, r \rangle + \langle w, r \rangle, \forall v, w, r \in V; \)
2. \( \langle av \rangle = a\langle v \rangle, \forall a \in \mathbb{R}, v \in V; \)
3. \( \langle v, w \rangle = \langle w, v \rangle, \forall v, w \in V; \)
4. $\langle v, v \rangle \leq 0, \forall v \in V$;

5. $\langle v, v \rangle = 0 \iff v = 0, \forall v \in V$;

A vector space $V$ together with an inner product $\langle \cdot, \cdot \rangle$ it is called inner product space.

**Hilbert space**

A Hilbert space $\mathcal{H}$ is a vector space with an inner product $\langle v, w \rangle$, and associated norm:

$$\|v\| = \sqrt{\langle v, v \rangle} \quad (2)$$

and metric, in which every Cauchy sequence is convergent.

**$L^2$ space**

An $L^2$ space is defined as the set of square integrable measurable functions $u : \Omega \to \mathbb{R}$ such that:

$$\langle u, u \rangle = \int_\Omega uu \, dx \quad (3)$$

being the norm $\|u\|_{L^2} = \sqrt{\langle u, u \rangle}, \forall u \in L^2(\Omega)$. Thus, the $L^2$ space forms a Hilbert space.
Part I

Mathematical modelling for distributed systems
Most of the processes of interest in the Food and Biotechnology industries involve variables, like concentrations and temperatures, or parameters which are dependent both on time and on space dimensions. According to this spatio-temporal nature, all these processes (from tubular reactors to moving boundary problems) can be consequently comprised into the class of the Distributed Parameter Systems (DPS).

Typically, DPS can be modelled by sets of coupled, usually non-linear, Partial Differential Equations (PDE’s), which can be derived from first-principles (i.e. mass, energy and momentum conservation laws). These characteristics make difficult to find analytical solutions, calling for the employment of numerical methods to solve the proposed problems.

Part I of this dissertation will be devoted to provide with an overview of those numerical techniques and their essential features. Besides, the computational tools that make use of them, and which will be further employed, will be also introduced.

Classical numerical methods employed to solve PDE’s, such as the Finite Element method (FDM) or Finite Difference method (FDM), will be outlined in Chapter 1. These methods make use of a domain discretisation to approximate the corresponding solution through local basis functions. Depending on the size of the discrete domain employed, and the number of equations to be solved, these methods might result computationally expensive, and unsuitable for certain real time applications.

As alternative, the Reduced Order Methods (ROM) arise as useful tool to lessen the dimensionality of the approximate solutions. By retaining only the slowest dynamics of the process, the essence of the system behaviour can be captured and
reproduced in a low-dimensional framework suitable for on-line applications. Chapter 2 will deal with these model reduction techniques, introducing two of the most representative ones: the Laplacian Spectral Decomposition (LSD) and the Proper Orthogonal Decomposition (POD). Likewise, particulars on the exploitation of the FEM matrices in their computational implementation will be given. To that purpose, the theoretical basis of those techniques will be presented together with an illustrative example of application.

Finally, the modelling of moving boundary problems, which constitute a particular case of distributed systems, will be introduce in Chapter 3. This class of systems exhibits time-varying spatial domains, where at least one boundary (maybe internal) is not known in advance, and whose position, consequently, must be determined as a function of time and space. Since few analytical solutions are available, the main challenges are related to develop accurate numerical approaches. By means of the solution of an illustrative example, the essential features of the selected numerical techniques to cope with moving boundaries will be introduced. In addition, the applicability of methods for model reduction in these challenging time-varying domains will be also analysed.
Introducción

A mayoría dos procesos de interese nas industrias alimentaria e biotecnolóxica involucran variables, tales como temperaturas e concentracións, ou parámetros, que dependen tanto da variable temporal coma espacial. De acordo con esta natureza espazo-temporal, todos estes procesos (desde reactores tubulares ata problemas de fronteira móbil) poden incluírse na categoría de sistemas distribuídos.

Os sistemas distribuídos adoitan modelarse empregando sistemas acoplados, normalmente non liñais, de ecuacións en derivadas parciais (EPD), que poden ser derivados a partires de primeiros principios (isto é, a partires das leis de conservación de enerxía, masa e momento). Estas características fan moi difícil, senón imposible, a obtención de solucións analíticas, facéndose necesario o emprego de métodos numéricos.

A Parte I desta disertación está adicada a ofrecer unha visión xeral destas técnicas numéricas e das súas características fundamentais. Ademais, vanse introducir tamén as ferramentas computacionais que permiten a súa implementación numérica.

Os métodos numéricos clásicos empregados para resolver ecuacións en derivadas parciais, tales como o Método dos Elementos Finitos (MEF) ou Método de Diferenzas Finitas (MDF), serán descritos no Capítulo 1. Estes métodos fan uso dunha discretización do dominio espacial para aproximar a correspondente solución do sistema mediante funcións base locais. Dependendo do tamaño do dominio discreto empregado, e do número de ecuacións que deben ser resoltas, estes métodos poden resultar computacionalmente custosos, e non axeitados para aplicacións en tempo real.

Como alternativa aos métodos clásicos xorden os Métodos de Orde Reducida
Introducción

(MOR), unha ferramenta moi útil para reducir a dimensionalidade dos sistemas. Retendo soamente as dinámicas máis lentas do sistema, pode capturarse a esencia do comportamento do mesmo, que será reproducida nun espazo de baixa dimensión adecuado para aplicacións en liña. O Capítulo 2 presenta estas técnicas de redución de modelos, introducindo dous dos seus máis representativos exemplos: a Descomposición Espectral do Laplaciano (LSD) e Descomposición Ortogonal Propia (POD). Do mesmo xeito, introducirase tamén o uso das matrices de Elementos Finitos na súa implementación computacional, todo mediante o emprego dun exemplo práctico de aplicación.

Por último, a modelaxe de problemas de fronteira móbil, que constitúen un caso particular de sistemas distribuídos, introducécese no Capítulo 3. Este tipo de sistemas presenta dominios espaciais que varían no tempo, nos que polo menos unha fronteira (pode que interior) é descoñecida, e da que deben ser determinadas, como unha función do tempo e do espazo, a súa posición e/ou velocidade. Como non hai moitas solucións analíticas dispoñibles, os principais retos están relacionados co desenvolvemento de esquemas numéricos precisos. Mediante a resolución dun exemplo ilustrativo van ser introducidas as características esenciais dos métodos empregados para resolver problemas de fronteira móbil. Tamén se vai analizar a aplicabilidade de métodos de redución do modelos en dominios variables dependentes do tempo.
1.1 Introduction

Distributed systems are those ones exhibiting input, output or even parameters which are functions of both temporal and spatial independent variables (Li and Qi, 2010).

Into this definition suit many physical and bio-chemical processes (i.e. tubular and fixed-bed reactors, heat exchangers, travelling waves, etc). Due to the inherent complexity of the different phenomena taking place in such processes, proper dynamical descriptions are required for system analysis, as well as for control design and optimisation applications. It is in this framework where mathematical modelling, together with numerical simulation, have been revealed as very necessary tools.

Typically, distributed systems are modelled by sets of coupled, usually non-linear, Partial Differential Equations (PDE’s), which can be derived from first-principles (i.e. mass, energy and momentum conservation laws). In its most general writing, a PDE could take the following form:

\[
d\frac{da}{dt} \frac{\partial u}{\partial t} = -\nabla \left( \nabla u \right) - \nabla \left( \nabla u \right) + f(u), \quad \forall u \in \Omega
\]  

(1.1)

where the temporal independent variable is defined as \( t \in [0, \infty) \) and it is considered the spatial domain \( \Omega \in \mathbb{R}^3 \). Additionally, mixed boundary conditions (any other type can be derived from this one) are imposed:

\[
\mathbf{n} \cdot \nabla u + qu = g, \quad \text{in} \quad \Gamma
\]  

(1.2)

In order to complete the system, the corresponding initial conditions must be defined, too. In this generic formulation, the term \( f(u) \) represents a non-linear function while
the coefficient \( da \) is considered to be a positive constant.

Distributed fields found as solution of PDE systems, such as the one above described, lie in infinite-dimensional Hilbert spaces. Therefore, they can be expanded in the form of a convergent Fourier series (Courant and Hilbert, 1953):

\[
 u(\zeta, t) = \sum_{i=1}^{\infty} m_i(t) \varphi_i(\zeta) 
\]

(1.3)

reading \( \{\varphi_i(\zeta)\}_{i=1}^{\infty} \) for the set of spatial dependent basis functions, while \( \{m_i(t)\}_{i=1}^{\infty} \) represents the temporal modes. This spatio-temporal separation will be exploited by the different numerical approaches aimed at solving PDE systems.

Classical numerical methods employed to solve PDE’s, such as the Finite Element method (FDM) or Finite Difference method (FDM), focus on the spatial variable and make use of a domain discretisation to approximate, through local basis functions \( \varphi \), the solution at the resulting finite set of \( N \) nodes or elements. Depending on the size of the discrete domain, and the number of equations to be solved, these methods might result computationally expensive, and unsuitable for certain fields, like real time optimization or predictive control, where the computing times are very important (Christofides, 2001).

As alternative, one can take advantage of the dissipative property of the distributed systems (Alonso and Ydstie, 2001) to lessen the dimensionality of the finite approximations. By working on the temporal model, and retaining only the slowest modes: \( \{m_i(t)\}_{i=1}^{neig} \) with \( neig \in \mathbb{N} \), those which most contribute to the system dynamics, and choosing, properly, global basis functions \( \phi \), it is possible to accurately approximate the field by those \( neig \) first terms of the expansion described in Eqn (1.3). This are the fundamentals of the so-called Reduced Order Methods (ROM), and among them, of the Laplacian Spectral Decomposition (LSD) and the Proper Orthogonal Decomposition (POD) (García et al., 2007).

The numerical methods aforementioned (FEM, FDM, LSD and POD) are the selected techniques to numerically solve the operational models subject of interest in the second part of this thesis.

Therefore, and although it is not the goal of this work to provide with an exhaustive description of any of those numerical techniques, an overview of their essential features is offered with an introductory purpose. Moreover, the computational tools that make use of them, and which will be further employed, are also introduced.

Next, it follows a brief presentation of the Finite Element Method (FEM) and the
Finite Difference Method (FDM), while Chapter 2 is devoted to the referred model reduction techniques, namely the Laplacian Spectral Decomposition (LSD) and the Proper Orthogonal Decomposition (POD).

1.2 Finite Difference Method

In this method, the derivatives in the PDE are approximated by difference quotients based on the values of the function on the points (nodes) of the discrete spatio-temporal domain (grid) (Li and Qi, 2010).

Generally, for a one-dimensional spatial domain $\Omega = [a,b]$, being the time interval $[0,t]$, the corresponding grid is defined as follows:

$$\Omega_{h,k} = \{ P_{i,j} = (a + ih, jk) | 0 \leq i \leq N, 0 \leq i \leq M \} \quad (1.4)$$

$$h = \frac{b-a}{N}; \quad k = \frac{t}{M}$$

The approximations are typically derived from the Taylor expansion around the nodes, exhibiting an order $O$ proportional to the distance between those, and thus dependent on the grid. If $u(x,t) \in C^{n+1}(\Omega)$, then:

$$u(x+h) \approx u(x) + \sum_{k=1}^{n} h^k \frac{\partial^k u}{\partial x^k} + O(||h||^{n+1}) \quad (1.5)$$

Substitution of each derivative for its corresponding approximation will lead to the associated discrete problem, which will consist of $N-1$ equations to be solved.

The Method of Lines (MOL) (Schiesser, 1991) is a special case of the Finite Difference method, in which only the partial derivatives in the spatial domain are replaced by their corresponding finite difference approximations (Li and Qi, 2010).

Actually, a computational tool based on the MOL (Wouwer et al., 2004) will be employed in this thesis in order to provide the finite differences operator in matrix form, as it will be illustrate in the context of the numerical application presented next.

1.2.1 FD application: a tubular reactor system

For the sake of illustration, the FD method will be next employed in order to solve the non-linear distributed system which describes the operation of a tubular reactor when an exothermic reaction \( A \rightarrow B \) \cite{Barje et al., 2013} is considered:

\[
\frac{\partial C}{\partial t}(x,t) = \alpha \frac{\partial^2 C}{\partial x^2}(x,t) - v_r \frac{\partial C}{\partial x}(x,t) - r(C,T), \quad \forall x \in (0, L)
\]  
(1.6)

\[
\frac{\partial T}{\partial t}(x,t) = \alpha \frac{\partial^2 T}{\partial x^2}(x,t) - v_r \frac{\partial T}{\partial x}(x,t) + \frac{4h}{\rho C_p d_r} (T_c - T(x,t)) + \frac{\Delta H}{\rho C_p} r(C,T), \quad \forall x \in (0, L)
\]  
(1.7)

It consists of a mass (Eqn (1.6)) and an energy balance (Eqn (1.7)), and exhibits a non-linearity in the form of a first-order kinetics reaction rate defined as function of the reactant concentration \( C(\text{mol}/\text{L}) \) and temperature \( T(\text{K}) \):

\[
r(C, T) = k_0 C \exp \left( -\frac{E}{RT} \right)
\]  
(1.8)

At the boundaries, Danckwerts conditions \cite{Laabissi et al., 2001} are imposed:

\[
\alpha \frac{\partial T}{\partial x}(0, t) = v_r (T(0, t) - T_{in}); \quad \alpha \frac{\partial C}{\partial x}(0, t) = v_r (C(0, t) - C_{in})
\]  
(1.9)

\[
\frac{\partial T}{\partial x}(L, t) = 0; \quad \frac{\partial C}{\partial x}(L, t) = 0
\]  
(1.10)

where \( T_{in} \) and \( C_{in} \) are the variables values for the inlet reactor. Finally, initially conditions are defined by:

\[
T(x, 0) = T_{in}; \quad C(x, 0) = C_{in};
\]  
(1.11)

Table 1.1 offers a complete description of the remaining model parameters.

<table>
<thead>
<tr>
<th>( \rho )</th>
<th>Density ([g/\text{L}])</th>
<th>( C_p )</th>
<th>Heat capacity ([\text{cal}/g\text{K}])</th>
</tr>
</thead>
<tbody>
<tr>
<td>( h )</td>
<td>Heat transfer coefficient ([\text{cal}/s\text{m}^2\text{K}])</td>
<td>( \alpha )</td>
<td>Diffusion coefficient ([\text{m}^2/\text{s}])</td>
</tr>
<tr>
<td>( d_r )</td>
<td>Reactor diameter ([\text{m}])</td>
<td>( \Delta H_r )</td>
<td>Reaction heat ([\text{kg}/\text{m}^3])</td>
</tr>
<tr>
<td>( v_r )</td>
<td>Flow velocity ([\text{m}/\text{s}])</td>
<td>( E )</td>
<td>Activation energy ([\text{cal}/\text{mol}])</td>
</tr>
<tr>
<td>( R )</td>
<td>Ideal Gas constant ([\text{cal}/\text{mol}\text{L}])</td>
<td>( L )</td>
<td>Reactor length ([\text{m}])</td>
</tr>
</tbody>
</table>

Table 1.1: Parameters for the tubular reactor model. The corresponding numerical values can be found in Barje et al. (2013).
First, the spatial domain is discretised by employing \( N = 31 \) nodes, which leads to \( \Omega = [0 : \delta x : L] \), where \( \delta x = (N - 1)/L \). Next, the spatial partial derivatives in the system are approximated, attending to its order, by the corresponding FD approximation.

Here, the function `matfd`, which is part of the free Matlab-based toolbox MatMOL (www.matmol.org), supplies the FD approximation, in matrix form, for each spatial derivative operator. The function requires only the spatial grid, the order of the derivative to be approximated and the information related to the stencil employed, i.e number of points, and to the desired scheme (backward, centered or forward).

For the example here presented, the first and second spatial derivatives are approximated by a 3 point stencil centered in space. Moreover, the integration in time has been carried out by a forward in time Matlab solver, the `ode15s`, for \( t = [0 : \delta t : 10] \). The time step \( \delta t \) is such that guarantees the stability of the FTCS\(^1\) approximation employed.

\[
\delta t \leq \frac{0.5 \delta x^2}{\alpha}
\]  

The results obtained are shown in Figure 1.1, where the distributed nature of the state variables clearly stands out.

![Figure 1.1: Evolution of the distributed state variables for the tubular reactor system: a) temperature \( T(K) \) and b) reactant concentration \( C(mol/L) \).](image)

\[\text{(a)}\] \[\text{(b)}\]

**1.3 The Finite Element Method**

Here it follows an outline of the Finite Element method in four steps, from the initial strong formulation to the final matrix form. In this section, and for the sake of

\(^1\text{Forward in Time Centered in Space}\)
simplicity, a one-dimensional domain $\Omega \in \mathbb{R}$ will be considered.

- **Strong formulation**: it is the set of governing PDE’s to be solved, with the corresponding boundary conditions, as described before by the general system formed by Eqns (1.1) and (1.2). Furthermore, the dependent variable is supposed to be regular enough, so $u \in H^2(\Omega)$.

- **Variational formulation**: The governing equation Eqn (1.1) is multiplied term by term by a test function $\Psi \in H^1(\Omega)$ and integrated over the spatial domain:

$$
\int_\Omega \Psi \frac{\partial u}{\partial t} dx = \int_\Omega \Psi \frac{\partial}{\partial x} \left( \frac{\partial u}{\partial x} \right) dx - \int_\Omega \Psi \frac{\partial (vu)}{\partial x} dx + \int_\Omega \Psi f(u) dx \quad (1.13)
$$

After applying Green’s theorem to downgrade the integral with the second derivative, and introducing the boundary conditions, the variational formulation will read:

$$
\int_\Omega \Psi \frac{\partial u}{\partial t} dx + \int_\Omega \frac{\partial \Psi}{\partial x} \frac{\partial (vu)}{\partial x} dx + \int_\Gamma \Psi q dx = \int_\Gamma \Psi g dx + \int_\Omega \Psi f(u) dx \quad (1.14)
$$

- **Galerkin’s method**: the spatial domain $\Omega$ is discretised into pieces, the so-called finite elements (segments for this 1D case), being the infinite-dimensional distributed variable $u(x,t)$ approximated in each of these pieces by the finite expression of the Fourier series presented in Eqn (1.3):

$$
u(x,t) \approx \sum_{i=1}^{N_e} U_i \varphi_i \quad (1.15)
$$

where $N_e$ are the number of nodes in the element, $U_i$ represents the values of the solution at the nodes and $\varphi_i$ notes the local basis functions.

- **Matrix form**: now, the approximated solution given by Eqn (1.15) is introduced into the variational formulation, where the test functions $\Psi$ are substituted by the local basis functions $\varphi_i$, too. Also at this step, the elements are assembled by considering continuity of the solution at each node. This will lead to:

$$
\text{da} \mathbf{M} \frac{\partial \mathbf{U}}{\partial t} + (\mathbf{D} \mathbf{M} + \mathbf{C} \mathbf{M} + q \mathbf{B} \mathbf{M}) \mathbf{U} = \mathbf{G} + \mathbf{F} \quad (1.16)
$$

where the FEM matrices are noted as follows: $\mathbf{M}$ is the mass matrix, $\mathbf{D} \mathbf{M}$ represents the diffusion matrix, $\mathbf{C} \mathbf{M}$ reads for the convection matrix, $\mathbf{B} \mathbf{M}$ and
1.3. The Finite Element Method

$G$ are the homogeneous matrix and the non-homogeneous vector of boundary conditions, respectively, and finally, $F$ accounts for the vector corresponding to the non-linear term. In addition, it must be mentioned that FEM matrices are defined in $\mathbb{R}^{N \times N}$ (i.e. so being square matrices), with $N$ noting the total number of nodes in the spatial grid.

For a deep and complete review of the method readers are referred to Reddy (1993), Zienkiewicz (2000) or García et al. (2007).

1.3.1 FEM application: the Kuramoto-Sivashinsky equation

The performance of the Finite Element technique is next exemplified in the solution of the one-dimensional Kuramoto-Sivashinsky (K-S) equation. This non-linear PDE is characterized by exhibiting a wealth of spatially and temporally non-trivial dynamical behaviour including chaos (Kevrekidis et al., 1990). It arises in a wide range of applications, such as reaction diffusion systems, long waves on the interface between two viscous fluids or thin hydrodynamics films (Uddin et al., 2009). Consequently, the K-S equation has been extensively used in the last decades as a model example in order to better understand systems with complex dynamics (Hyman and Nicoalenco, 1986; Armbuster et al., 1989; Kevrekidis et al., 1990; Smyrlis and Papageorgiou, 1996; Armaou and Christofides, 2000, 2005).

In its generalized form (Khater and Temsah, 2008), the Kuramoto-Sivashinsky equation reads as follows:

$$\frac{\partial u(x, t)}{\partial t} = -u(x, t) \frac{\partial u}{\partial x}(x, t) - a \frac{\partial^2 u}{\partial x^2}(x, t) - b \frac{\partial^3 u}{\partial x^3}(x, t) - c \frac{\partial^4 u}{\partial x^4}(x, t)$$

(1.17)

where the values of the coefficients $a$, $b$ and $c$ determine the nature of the system.

Exact solutions for the K-S equation have been reported by Xu et al. (2006) when the following set of coefficients was employed: $a = 1$ and $b = 4$, $c = 1$. In this case, the solution exhibited the form of a travelling wave with constant shape.

On the other hand, when coefficients $a$ and $c$ are both positive, being $b = 0$, the linear terms in Eqn (1.17) describe a balance between long-wave instability and short-wave stability, while the non-linear term provides a mechanism for energy transfer between wave modes. This results into a chaotic behaviour if periodic boundary conditions are considered (Xu et al., 2006).

Due to the existence of a known solution to compare with, the first case above mentioned has been chosen in order to illustrate step by step the application of the
Chapter 1

Finite Element Method. Results obtained for the chaotic system by means of the same numerical procedure will be presented next.

Case 1: K-S with exact solution

Consider the particular form of Eqn (1.17) defined by coefficients $a = 1$, $b = 4$ and $c = 1$, with initial conditions:

$$u(x, 0) = C + 9 - 15[tanh (k (x - x_0)) + tanh^2 (k (x - x_0)) - tanh^3 (k (x - x_0))]$$

which discloses an exact solution given by Xu et al. (2006):

$$u(x, t) = C + 9 - 15[tanh (k (x - Ct - x_0)) + tanh^2 (k (x - Ct - x_0)) - tanh^3 (k (x - Ct - x_0))]$$

In order to apply the FE method, the spatial domain $\Omega = [-L, L]$ is homogeneously discretised by employing $N = 128$ points, what implies $N - 1$ Lagrange P1 finite elements in the form of segments, since a one-dimensional geometry is being considered.

Figure 1.2: Distributed exact solution for the Kuramoto-Sivashinsky equation obtained for $t = [0 : 0.001 : 4]$, and $x \in [-30, 30]$. 
The variational formulation for the problem before described reads:

\[
\frac{\partial}{\partial t} \int_{\Omega} \psi u(x,t) \, dx = - \int_{\Omega} \psi u(x,t) \frac{\partial u}{\partial x}(x,t) \, dx - \int_{\Omega} \psi \frac{\partial^2 u}{\partial x^2}(x,t) \, dx - 4 \int_{\Omega} \psi \frac{\partial^3 u}{\partial x^3}(x,t) \, dx - \int_{\Omega} \psi \frac{\partial^4 u}{\partial x^4}(x,t) \, dx
\]

for any test function \( \psi \in H^1(\Omega) \). As stated in García et al. (2007), there exist algebraic equivalences for efficient numerical computation of integrals and spatial derivative by means of the FEM matrices. Thus, after substitution of the test functions by the local basis functions \( \varphi_i(x) \), employing the FEM matrices also for integrating/differentiating and assembling, and noting by \( U \) the discrete state variable, the resulting ODE system is obtained:

\[
\frac{\partial U}{\partial t} = - U M M^{-1} (CM) U + M M^{-1} (DM + BM) U + 4 M M^{-1} (CM) MM^{-1} (DM + BM) U + M M^{-1} (DM + BM) MM^{-1} (DM + BM) U
\]

Since this is a simple 1D domain, the MatMOL toolbox can be employed again in order to obtain the FEM matrices through its function \textit{matfem}. The inputs to be provided are the spatial discretisation and the type of boundary conditions. If geometries more complex were considered, other FEM oriented commercial packages, like COMSOL Multiphysics (COMSOL AB, 2008), can be used to extract the corresponding set of matrices. The solver \textit{ode15s} from Matlab has been chosen, once more, for time integration of the new ODE system.
Results are shown in Figure 1.3 in comparison to the exact solution, confirming the accuracy, even for complex DPS like the K-S equation, of the finite-dimensional approximation given by the FEM (computed values for absolute errors of $e_{abs}^{max} = 0.18$ and $\bar{e}_{abs} = 0.01$).

Figure 1.3: Comparison between FEM and exact solution time profiles for the Kuramoto-Sivashinsky equation with $C = 6$, $k = 0.5$ and $x_0 = -10$.

**Case 2: K-S with chaotic solution**

Once the suitability of the FEM method to deal with such complex system has been showed, the FEM-based numerical solving of the K-S chaotic form is now tackled.

For this second case, where coefficients are given by $a = 1$, $b = 0$ and $c = 1$, a Gaussian initial condition will be considered (Uddin et al., 2009):

$$u(x, 0) = e^{-x^2}$$

(1.21)

Together with periodic boundary conditions:

$$u(-L, t) = u(L, t) = 0$$

(1.22)

The spatial domain is the same one already defined for the first case, with $L = 30$, as well as the number of nodes ($N = 128$). Figure 1.4 shows the distributed spatio-temporal solution obtained. It can be seen the exponential growing in time of the
1.3. The Finite Element Method

small perturbations introduced by the initial condition. Consequently, predictability of the system is lost, revealing the chaotic nature of the system (Brummitt and Sprott, 2009). Note the differences with respect to Figure 1.2, where a soliton solution was depicted.

![Figure 1.4](image)

Figure 1.4: Distributed solution for the chaotic Kuramoto-Sivashinsky system obtained by FEM with $t = [0 : 0.001 : 20]$. 
2.1 Introduction

The realistic description, in mathematical terms, of a distributed system might involve a high number of Partial differential Equations, usually coupled and with non-linear terms.

When dealing with these large-scale models, the use of the most widespread methods of solving PDE systems, such as FEM or FDM, entails the handling of a too large number of equations, which yet raises orders of magnitude when 2D or 3D geometries are considered. Such large-order and complex models would be unsuitable if their final purpose is being employed within the real-time applications framework, namely on-line optimisation or model-based control.

A simplification of the original model arises as an interesting option then, but some important guidelines must be kept in mind. The simplified version should preserve the same system properties (e.g. stability or passivity), and it should also preserve the same order of accuracy while exhibits computational efficiency.

The proposed simplification, namely model reduction, can be done in two different ways (Theodoropoulos, 2011): physically, attending to the insights of the system behaviour defined e.g. by the thermophysical and geometric properties, which would either lead to the identification of the involved time-scales, or defined suitable dimensionless transformations, etc.; mathematically, following diverse numerical methods headed to lessen the high dimensionality of the system by, e.g. capturing the relevant dynamics of the full model and further projection of the original system onto a lower dimension subspace.

Attending to the mathematical approach, model reduction techniques could be
referred to the set of numerical techniques aimed at approximating the classic methods of solving PDE systems, in order to obtain and accurate solution while reducing the computational burdens and preserving the main features of the original model.

Within this group of model reduction techniques the theory of Approximate Inertial Manifolds (Foias et al., 1988; Debusche and Marion, 1992), the Truncation Balance Method (Tombs and Postlethwaite, 1987), the Hankel-norm Reduction (Glover, 1984) or the Proper Orthogonal Decomposition (Sirovich, 1987; Holmes et al., 1996) can be found. An exhaustive review of those and some other ROMs is available in Antoulas (2005) or Schilders et al. (2008).

Accordingly with the advantages offered, reduced order models have been widely employed in the most diverse industrial problems, from chemical reactors (Alonso et al., 2004; Garcia et al., 2008; Lopez-Quiroga et al., 2010) to food processing (Balsa-Canto et al., 2002), including control applications (Ito and Kunisch, 2006; Garcia et al., 2012), successfully obtaining, in all those cases, the desired simplified and low-order description of the process dynamics.

The scope of this chapter is to provide a brief introduction to the reduced order methods employed in obtaining part of the results presented in this thesis, as well as to give particulars on the exploitation of the FEM matrices in their computational implementation. To that purpose, the theoretic basis of those techniques, together with an illustrative example of application, are presented. Section 2.3 tackles the Proper Orthogonal Decomposition (POD), while the Laplacian Spectral Decomposition (LSD) will be addressed in Section 2.4.

### 2.2 Model reduction techniques

The two techniques employed along this work to effectively obtain low-dimensional but accurate dynamic models are based on an eigenfunction approach. In this approach, each basis function \( \phi_i(\vec{\zeta}) \) of the system is computed as the solution of the following eigenvalue problem:

\[
\int_{\Omega} K(\vec{\zeta}, \vec{\zeta}') \phi_i(\vec{\zeta}) d\vec{\zeta}' = \lambda_i \phi_i(\vec{\zeta})
\]  

(2.1)

noting \( \Omega \) the spatial domain where the PDE system is formulated in, and being \( \lambda_i \) the eigenvalue associated to eigenfunction \( \phi_i \). The resulting spatial dependent basis functions are globally defined, and they also satisfy the imposed boundary conditions.
2.2. Model reduction techniques

It must be taken into account too that, if the kernel $K$ in Eqn (2.1) is symmetric, then the corresponding set of eigenfunctions will be orthonormal (Courant and Hilbert, 1953). Therefore,

$$<\phi_i, \phi_j>_{\Omega} = \begin{cases} 1 & \text{if } i = j \\ 0 & \text{if } i \neq j \end{cases}$$

(2.2)

Another point worthy of mention regarding Eqn (2.1) and dissipative nature of processes, is the existence of a spectral gap of characteristic time scales separating slow and fast components of the system dynamics (Armaou et al., 2005).

As already commented in Section 1.1, because the fast modes contribute little to the whole system, only the slowest ones, those which most contribute to the system dynamics, will be retained by model reduction techniques. Consequently, the infinite series in Eqn (1.3) can be truncated, with arbitrarily accuracy by a small number of terms (Alonso and Ydstie, 2001), which will be noted by $\text{neig}$:

$$u(\mathbf{\zeta}, t) \approx \hat{u}(\mathbf{\zeta}, t) = \sum_{i=1}^{\text{neig}} m_i(t) \phi_i(\mathbf{\zeta})$$

(2.3)

where $\{\phi_i(\mathbf{\zeta})\}_{i=1}^{\text{neig}}$ is the set of orthogonal basis functions computed through Eqn (2.1), while the mode set $\{m_i(t)\}_{i=1}^{\text{neig}}$ comprises those most representative temporal modes of the system.

In addition, the summation $\sum_{i=1}^{\text{neig}} \lambda_i$ of the associated eigenvalues will give an idea of the energy captured by the approximation, and also of its quality: the more the energy captured, the better the approximation provided by the ROM.

It is the nature of the kernel $K$ what determines different sets of basis functions (Alonso et al., 2004) and, related to them, different reduction methods, having among those:

- the Proper Orthogonal Decomposition (POD), method with a two-point correlation kernel defined by:

$$K(\mathbf{\zeta}, \mathbf{\zeta}') = \lim_{\theta \to \infty} \int_0^\theta u(t, \mathbf{\zeta}) u(t, \mathbf{\zeta}') dt$$

(2.4)

where $u(t, \mathbf{\zeta})$ represents the value of the field in the time interval $[0, \theta]$.

- the Laplacian Spectral Decomposition (LSD), where the kernel is the Green’s function associated with the Laplacian operator.
Both methods will be next described.

### 2.3 Proper Orthogonal Decomposition

The computation of the basis functions in the POD technique requires to collect a database of measurements carefully selected so they capture the relevant dynamics of the system. These data, the so-called snapshots, represent the state variables, or a perturbed version of them, at a finite number of spatial nodes \( N \) and at different process times \( t_j \), being noted by \( u_j(\vec{\zeta}), j = 1, ..., l \). Either direct measurements or numerical simulation can be employed in order to assemble this set of system samples.

On the suitability of the snapshots collection relies the goodness of the low-dimensional representation obtained, since the kernel, in terms of this finite set of measurements, can be defined as:

\[
K(\vec{\zeta}, \vec{\zeta}') = \frac{1}{l} \sum_{i=j}^{l} u_j(\vec{\zeta}, t_j) u_j(\vec{\zeta}', t_j) \tag{2.5}
\]

and the dynamical behaviour of the system, also with regard to the effects of nonlinearities and/or perturbations, is expected to be enclosed therein.

Aiming at obtaining the discrete version of Eqn (2.1), the equivalence for integrals expressed as a FEM matrix product, see Table 1.2, is here employed, leading to:

\[
KMM\Phi_i = \lambda_i \Phi_i, \quad i = 1, ..., neig \tag{2.6}
\]

where the kernel is now redefined when considering the \( U_j \) vector in \( \mathbb{R}^N \) of \( l \) snapshots:

\[
K = \frac{1}{l} \sum_{j=1}^{l} U_j U_j^T \tag{2.7}
\]

and \( \Phi_i \in \mathbb{R}^N \) represents the discrete counterpart of the eigenfunction \( \phi_i(\vec{\zeta}) \).

Back to Eqn (2.3), the criterion for determining the number of basis functions, \( neig \), employed for approximating the field relies on the energy captured by its corresponding eigenvalues \( \lambda_i \) (Sirovich, 1987), which is measured by:

\[
E = 100 \times \frac{\sum_{i=1}^{neig} \lambda_i}{\sum_{i=1}^{l} \lambda_i} \tag{2.8}
\]
Finally, the PDE system, including boundary and initial conditions as well as non-linear terms, must be projected onto the selected \( \text{neig} \) POD basis. This results into a dimensionally reduced set of ODEs to be solved. The complete numerical procedure will be next described.

### 2.3.1 POD-based solution for the Kuramoto-Sivashinsky equation

Aiming at providing an illustrative application of the POD method, the one-dimensional Kuramoto-Sivashinsky (K-S) system already presented in Section 1.3.1 Case 1, is here recovered.

\[
\frac{\partial u}{\partial t}(x, t) = f(u, \frac{\partial u}{\partial x}, \frac{\partial^2 u}{\partial x^2}, \frac{\partial^3 u}{\partial x^3}, \frac{\partial^4 u}{\partial x^4}),
\]

\[
u(-L, t) = u(L, t),
\]

\[
u(x, 0) = u_0(x)
\]

which reads as the compact notation for the K-S system, since the RHS in Eqn (1.17) is here generically expressed as a function of the field \( u \) and its spatial derivatives.

When the POD is the chosen method for model reduction, the first step to take is to collect a proper, namely a rich and uncorrelated, set of snapshots. For this example, this is done by numerical simulation of the FEM model (see Chapter 1) and results were recorded with a time interval \( \delta t = 0.02 \; \text{s} \).

The snapshots were provided as input, together with the FEM matrices of the system and the spatial discretisation, to the function \text{matpod} (www.matmol.org). This function also requires to specify the value of energy to be captured by the reduced system, and it returns as outputs the \( \text{neig} \) discrete counterparts of the POD basis, \( \Phi_i \), and eigenvalues \( \lambda_i \) which fulfils such energy requirement.

After this, it follows Galerkin projection of system (2.9), which consists of multiplying those equations, term by term, by each basis function and, after then, integrating over the spatial domain \( \Omega \). Therefore, the spatial and time derivative operators for the approximated solution \( \tilde{u}(x, t) \), according Eqn (2.3), will read:

\[
\frac{\partial u}{\partial t} \approx \left( \sum_{i=1}^{\text{neig}} m_i \phi_i \right) = \sum_{i=1}^{\text{neig}} \frac{\partial m_i}{\partial t} \int_{\Omega} \phi_i^T \phi_i dx = \frac{dm_i}{dt}
\]
after applying orthonormality (property defined in Eqn (2.2)).

\[
\frac{\partial^2 u}{\partial x^2} \approx \frac{\partial^2}{\partial x^2} \left( \sum_{i=1}^{\text{neig}} m_i(t) \phi_i(t) \right) = \sum_{i=1}^{\text{neig}} m_i(t) \int_{\Omega} \phi_i^T \frac{\partial^2 \phi_i}{\partial x^2} dx = \tag{2.11}
\]

\[
= \sum_{i=1}^{\text{neig}} \left( -\Phi_i^T (D\mathcal{M} + B\mathcal{M}) \Phi_i \right) m_i
\]

where the correspondences presented in Table 1.2 has been employed again, as well as for the remaining derivatives:

\[
\frac{\partial^3 u}{\partial x^3} \approx \frac{\partial^3}{\partial x^3} \left( \sum_{i=1}^{\text{neig}} m_i(t) \phi_i(t) \right) = \sum_{i=1}^{\text{neig}} m_i(t) \int_{\Omega} \phi_i^T \frac{\partial^3 \phi_i}{\partial x^3} dx = \tag{2.12}
\]

\[
= \sum_{i=1}^{\text{neig}} \left( -\Phi_i^T (C\mathcal{M} \mathcal{M}^{-1} (D\mathcal{M} + B\mathcal{M}) \Phi_i \right) m_i
\]

\[
\frac{\partial^4 u}{\partial x^4} \approx \frac{\partial^4}{\partial x^4} \left( \sum_{i=1}^{\text{neig}} m_i(t) \phi_i(t) \right) = \sum_{i=1}^{\text{neig}} m_i(t) \int_{\Omega} \phi_i^T \frac{\partial^4 \phi_i}{\partial x^4} dx = \tag{2.13}
\]

\[
= \sum_{i=1}^{\text{neig}} \left( \Phi_i^T (D\mathcal{M} + B\mathcal{M}) \mathcal{M}^{-1} (D\mathcal{M} + B\mathcal{M}) \Phi_i \right) m_i
\]

Finally, the projection of the non-linear term \( g = u \frac{\partial u}{\partial x} \) will be tackled following the strategy first proposed in Vilas (2008):

\[
u \frac{\partial u}{\partial x} \approx \int_{\Omega} \phi_i^T g(x,t) = \Phi_i^T \mathcal{M} G, \quad i = 1, ..., \text{neig} \tag{2.14}
\]

being \( G \) the discrete version of \( g(x,t) \). The resulting reduced ODE system is next described:

\[
\frac{dm}{dt} = -\Phi^T \mathcal{M} G + \mathcal{A} m \tag{2.15}
\]

with \( m = [m_1, ..., m_{\text{neig}}]^T \), and \( \Phi = [\Phi_1, ..., \Phi_{\text{neig}}] \). Besides, \( \mathcal{A} \) represents the addition of the spatial derivatives projected onto the POD basis, as derived in Eqns (2.11)-(2.13):

\[
\mathcal{A} = \Phi^T (D\mathcal{M} + B\mathcal{M}) \Phi + \Phi^T C\mathcal{M} \mathcal{M}^{-1} (D\mathcal{M} + B\mathcal{M}) \Phi + \tag{2.16}
\]

\[
+ \Phi^T (D\mathcal{M} + B\mathcal{M}) \mathcal{M}^{-1} (D\mathcal{M} + B\mathcal{M}) \Phi \tag{2.17}
\]
2.3. Proper Orthogonal Decomposition

Figure 2.1: The first three POD basis functions computed through function matpod for the Kuramoto-Sivashinsky equation.

The initial condition for this system is also obtained by projection of the original initial value, which results into:

$$m(0) = \Phi^T \mathcal{M} \mathcal{M}^\dagger m(0) \quad (2.18)$$

The system formed by Eqns (2.15) and (2.18) is numerically solved by means of the Matlab solver ode15s for $t = [0, 4]$, with time step $\delta t = 0.1$. The evolution in time for the first three modes is shown in Figure 2.2. The approximated field, $\tilde{u}(x, t)$, can be easily recovered by computing:

$$\tilde{u}(x, t) = \sum_{i=1}^{neig} \Phi_i m_i \quad (2.19)$$

Three different time profiles obtained for $\tilde{u}(x, t)$ are depicted in Figure 2.3, and compared to the FEM system solutions at the same times of simulation. A very good agreement between them can be observed, being the maximum absolute error $e_{abs}^{max} = 0.41$ and the mean absolute error $\bar{e}_{abs} = 0.02$.

The number of modes employed to obtain this approximation is $neig = 31$, which implies a reduction of up to 87% in the number of degrees of freedom solved when compared with the full model, namely the FEM model, which solves a system of 256 ODE’s.
2.4 Laplacian Spectral Decomposition

According to Courant and Hilbert (1953), solving the eigenvalue problem formulated in Eqn (2.1) together with homogeneous boundary conditions, equates with solving the following spectral problem:

$$\Delta \phi_i(\vec{\zeta}) = -\lambda_i \phi_i(\vec{\zeta})$$  \hspace{1cm} (2.20)

Therefore, the basis functions employed by the LSD technique are those computed as the solution of Eqn (2.20), when the proper boundary conditions are imposed. For those systems with non-homogeneous boundary conditions, the transformations proposed by Courant and Hilbert (1953) and Balsa-Canto et al. (2004a) will be applied in order to meet the aforementioned requirement.

The equivalences before presented in Table 1.2 will be employed again to obtain the discrete version of the eigenproblem defined by Eqn (2.20), which reads:

$$\mathcal{M} \mathcal{M}^{-1} (\mathcal{D} \mathcal{M} + \mathcal{B} \mathcal{M}) \Phi_i = -\lambda_i \Phi_i$$ \hspace{1cm} (2.21)

where $\Phi_i \in \mathbb{R}^N$ refers to the discrete version of the basis functions, which are supposed to form an orthonormal set in $L^2(\Omega)$. Moreover, it must be noted that they only depend on the spatial grid.

The application of the LSD method from here on is analogous to what has been
already described in 2.3, since next steps involved projection of the PDE onto the computed discrete basis functions to obtain the corresponding low-dimensional ODE system. For the sake of comparison, the LSD method will be employed to solve the same illustrative examples next.

2.4.1 LSD-based solution for the Kuramoto-Sivashinsky equation

Although the boundary conditions specified for the so-called exact form of the K-S equation exhibit a complex time-dependent form, as described in Section 1.3.1 Case 1, in practice its values are constant and equal to zero in the time horizon of the simulation, as can be seen in Figure 1.3. Thus, they can be approximated by the following expressions without affecting the system nature:

\[ u(-L, t) = u(L, t) = 0 \]  \hspace{1cm} (2.22)

Since they are Dirichlet homogeneous conditions, LSD method can be directly applied without performing any transformation of the equations set.

Getting into the numerical procedure, the *matlsd* function (by *matmol* toolbox) will be employed to compute the discretised basis functions, \( \Phi_i \), and eigenvalues \( \lambda_i \), by solving the problem formalised in Eqn (2.21) when the homogeneous boundary
Chapter 2

conditions are imposed: Next step is projection of the PDE system onto the LSD basis. Again, term by term:

$$\frac{\partial u}{\partial t} \approx \frac{d m_i}{dt}$$  \hspace{1cm} (2.23)

The derivation of this equivalence can be followed in Eqn (2.10).

$$\frac{\partial^2 u}{\partial x^2} \approx \sum_{i=1}^{\text{neig}} m_i \int_{\Omega} \phi_i \frac{\partial^2 \phi_i}{\partial x^2} = -\lambda_i m_i, \hspace{0.5cm} i = 1, ..., \text{neig}$$  \hspace{1cm} (2.24)

by employing the equality in Eqn (2.20) and the correspondences in Table 1.2.

$$\frac{\partial^3 u}{\partial x^3} \approx \sum_{i=1}^{\text{neig}} m_i(t) \int_{\Omega} \phi_i \frac{\partial^3 \phi_i}{\partial x^3} dx = \sum_{i=1}^{\text{neig}} (\Phi_i^T \mathcal{CM} \lambda_i \Phi_i) m_i, \hspace{0.5cm} i = 1, ..., \text{neig}$$  \hspace{1cm} (2.25)

where again the FEM matrices are employed, as well as Eqn (2.20). This equation is also used to compose the fourth derivative projection:

$$\frac{\partial^4 u}{\partial x^4} \approx \sum_{i=1}^{\text{neig}} m_i(t) \int_{\Omega} \phi_i \frac{\partial^4 \phi_i}{\partial x^4} dx = \sum_{i=1}^{\text{neig}} (\Phi_i^T \mathcal{C} \lambda_i \Phi_i^T) m_i$$  \hspace{1cm} (2.26)

The projection of the non-linear term is carried out in the same manner than it was done before for the POD technique, so Eqn (2.14) will be valid here, too, as well as the initial conditions defined in Eqn (2.18). The resulting reduced ODE system
2.4. Laplacian Spectral Decomposition

presents the same structure than Eqn (2.16), but now the operator $\mathcal{A}$ will read:

$$
\mathcal{A} = -\Lambda + \Phi^T C M \Lambda \Phi + \Phi^T \Lambda \Lambda \Phi
$$

where $\Lambda \in \mathbb{R}^{neig \times neig}$ is a diagonal matrix with the eigenvalues $\lambda_i$ as elements.

The Matlab solver ode15s has been again chosen to computationally solve the above system for $t = [0 : 0.1 : 4]$. Figure 2.5 shows the evolution in time of the first three modes of the reduced system.

![Figure 2.5: Evolution in time of the first three modes associated to the LSD-based low-dimensional system.](image)

Figure 2.5: Evolution in time of the first three modes associated to the LSD-based low-dimensional system.

![Figure 2.6: Comparison at different simulation times between the full model solution (solid) of the K-S equation computed by FEM and the LSD results (asterisks).](image)

Figure 2.6: Comparison at different simulation times between the full model solution (solid) of the K-S equation computed by FEM and the LSD results (asterisks).

The approximation $\tilde{u}(x, t)$ is recovered by means of Eqn (2.19). The results obtained, in terms of $\tilde{u}$, are plotted in Figure 2.6 together with those other ones corresponding to the FEM solution (full model). As can be seen, a good reproduction
of the systems dynamics is achieved by the LSD-based reduced order model, with a maximum absolute error of $e_{\text{max}}^\text{abs} = 0.41$ and a mean absolute error $\bar{e}_{\text{abs}} = 0.02$.

It must be noted that the LSD system solves only 56 degrees of freedom (neig=56), from the original number of 256 (nodes of the spatial grid) which are solved for the full model. This represents a reduction of around the 78% in the number of ODE’s to be solved, which also translates into a reduction of the related computation times.

### 2.5 ROM comparison

To end up the chapter, the performance of the POD and the LSD methods when employed to obtain the reduced model of the same system (the Kuramoto-Sivashinsky equation) is compared.

As shown in Table 2.1, the LSD technique needs 56 modes to provide and approximation displaying the same accuracy than the solution provided by the POD with only 31 modes. Clearly, from a comparative point of view, results show the higher power reduction exhibited by the POD technique. This is not unexpected, though, since it is well known the optimality, in a least square sense, of this technique (Sirovich, 1987). Nevertheless, there will be applications for which the LSD will be preferable, either due to the system structure or because of the difficulty to obtain a suitable set of snapshots. The Kuramoto-Sivashinsky equation itself serves as example of this assertion. When the chaotic form of the K-S (1.3.1) is considered for model reduction, the chaos exhibited by the K-S evolutionary solution makes difficult to capture the

<table>
<thead>
<tr>
<th></th>
<th>LSD $\text{neig} = 31$</th>
<th>POD $\text{neig} = 56$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Exact</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$e_{\text{max}}^\text{abs}$</td>
<td>16.07</td>
<td>0.38</td>
</tr>
<tr>
<td>$\bar{e}_{\text{abs}}$</td>
<td>0.59</td>
<td>0.020</td>
</tr>
<tr>
<td><strong>Chaotic</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$e_{\text{max}}^\text{abs}$</td>
<td>0.23</td>
<td>0.18</td>
</tr>
<tr>
<td>$\bar{e}_{\text{abs}}$</td>
<td>0.002</td>
<td>0.14</td>
</tr>
</tbody>
</table>

Table 2.1: Comparison between the LSD and POD methods for the Kuramoto-Sivashinky equation when $N = 256$, $t = [0 : 0.001 : 4]$. In blue, the best results for both solutions when 31 modes are used.
2.5. ROM comparison

system dynamics into a representative set of snapshots.

In addition, LSD might better exploit the system structure, taking advantage of the equivalence expressed in Eqn (2.20). Consequently, for this particular case, the LSD offers a better performance in terms of power reduction, as reflected in Table 2.1.
Chapter 2
3.1 Introduction

A particular case of distributed systems are those exhibiting time-varying spatial domains, where at least one boundary (maybe internal) is not known in advance, and whose position, consequently, must be determined as a function of time and space.

Among the wide range of applications comprised in the above description one can find separation processes (Mascarenhas et al., 1997), crack propagation (Stolarska et al., 2001), crystal growth (Ng et al., 2013), casting (Bermúdez and Otero, 2006) or laser ablation (Mitchell and Vynnycky, 2012). Generally, the mathematical formulation of such processes is referred to as moving boundary problems, while the denomination Stefan problem is specifically employed to identify problems involving moving boundaries where a phase-change occurs.

Due to the dependence of the moving front on the initial and boundary data, the superposition principle cannot be valid, thus making the problem non-linear (Landau, 1950; Fazio, 2013), and complicating its solution, either analytically or numerically (Liu, 1997). Since few analytical solutions are available, the main challenges are related to develop accurate numerical approaches.

As regards these numerical techniques, they are typically classified into three groups (Crank, 1987): front-tracking, front-fixing and fixed-domain methods. The first two approaches aim to describe and to directly handle the moving boundary, which position is computed at each time step. While the front-tracking schemes employ adaptive meshes or special formulae to compute the derivatives in the vicinity of the moving front, the front-fixing techniques make use of variable transformations in order to locate the moving interface on a pre-selected point of the spatial grid.
On the other hand, the fixed-domain methods rely on a reformulation of the problem such that the resulting system is defined over the whole workspace (Crank, 1987). Therefore, the position of the moving boundary is no longer considered, and its location is determined *a posteriori* (Furzeland, 1980; Crank, 1987). Excellent surveys on numerical methods for solving moving boundary problems can be found in literature (Crank, 1987; Gupta, 2003; Furzeland, 1980; Hu and Argyropoulos, 1996; Caldwell and Kwan, 2004; Javierre et al., 2006).

With respect to model reduction, the application of reduced order methods is straightforward in time-dependent spatial domains if it is considered in association with *front-fixing* (Armaou and Christofides, 2001a) or fixed domain schemes. More difficulties emerge when projection-based reduced order methods are meant to be combined with front-tracking techniques. Although, some efforts have been made recently in that sense (Izadi and Dubljevic, 2013; Stankiewicz et al., 2013).

This chapter provides a description of those numerical techniques, devoted to cope with moving boundary problems, which will be put to use in subsequent chapters. By means of the solution of an illustrative example, the essential features of three selected numerical techniques will be reported and compared. In addition, the applicability of methods for model reduction in these challenging time-varying domains will be also analysed.

### 3.2 Problem description: an example

Consider a one-dimensional domain occupied by a material which can exhibit both liquid and solid states, depending on temperature, as depicted in Figure 3.1.

Initially, both phases coexist, separated by the moving phase change front, \( S(t) \). The resulting problem, a two-phase Stefan-type problem, defines the freezing of the slab. Mathematically, it can be described as follows.

The heat flow in each region is governed by the corresponding Fourier equations:

\[
\rho_s C_p s \frac{\partial T_s}{\partial t} = k_s \frac{\partial^2 T_s}{\partial x^2}, \quad 0 \leq x < S(t)
\]

(3.1)

\[
\rho_l C_p l \frac{\partial T_l}{\partial t} = k_l \frac{\partial^2 T_l}{\partial x^2}, \quad x \geq S(t)
\]

(3.2)

where subscript \( s \) refers to the solid region while the subscript \( l \) is used to define the liquid phase. The thermophysical properties of both states are noted by \( \rho \) for the
3.2. Problem description: an example

density, $C_p$ for the heat capacity and $k$ for the heat conductivity.

![Diagram of spatial domain](image)

Figure 3.1: Spatial domain considered for a one-dimensional phase change problem.

In general, in a moving boundary problem governed by a second order partial differential equation, two boundary conditions are necessary on the front.

The first one, the so-called Stefan condition, defines the velocity of the interface, $w(t)$:

$$-k \frac{\partial T_l}{\partial x} \bigg|_{S(t)+} + k \frac{\partial T_s}{\partial x} \bigg|_{S(t)-} = \rho_s \Delta H w(t), \quad x = S(t)$$

being $\Delta H$ the latent heat. Note that the front velocity is formally:

$$w(t) = \frac{\partial S(t)}{\partial t}$$

The other condition is commonly related to the continuity of the temperature field across the moving boundary:

$$T_s(S(t), t) = T_l(S(t), t) = T_f$$

reading $T_f$ for the constant temperature at which the material freezes. The temperature at the exterior boundaries is given by:

$$T_s(0, t) = T_c(< T_f)$$

$$T_l(L, t) = T_L(> T_f)$$

Finally, the system is closed by the suitable initial conditions:

$$T(x, 0) = T_{ini}(x)$$
3.2.1 The Stefan condition

One of the main characteristics of the moving boundary problems is the additional condition required to describe the motion of the front, namely the Stefan condition (3.3). As shown next, it can be easily derived from the heat balance across the interface (Alexiades and Solomon, 1993):

\[
\left[\rho e\right]_n v_n = \left[\nabla q\right]_n \hat{n}
\]  

(3.9)

where \(v_n\) represents the normal velocity of the moving front and the term \(\left[\rho e\right]_n\) accounts for the energy jump caused by the phase change. Since for the chosen example both materials are supposed to be of the same density, and taking into account that the energy involved in the phase change is no other but the corresponding latent heat \(\Delta H\), it follows:

\[
\left[\rho e\right]_n v_n = \Delta H \rho_s v_n
\]  

(3.10)

The other term in Eqn (3.9) refers to the normal heat fluxes across the interface. Considering the Fourier law leads to:

\[
\left[\nabla q\right]_n \hat{n} = -k_l \nabla T_l \bigg|_{S^+} \hat{n}^l + k_s \nabla T_s \bigg|_{S^+} \hat{n}^s
\]  

(3.11)

Thus, the heat balance across the moving boundary can be now rewritten as:

\[
\Delta H \rho_s v_n = -k_l \nabla T_l \bigg|_{S^+} \hat{n}^l + k_s \nabla T_s \bigg|_{S^+} \hat{n}^s
\]  

(3.12)

For a one-dimensional case, the final expression obtained will be:

\[
\Delta H \rho_s \frac{\partial S(t)}{\partial t} = -k_l \frac{\partial T_l}{\partial x} \bigg|_{S(t)^+} + k_s \frac{\partial T_s}{\partial x} \bigg|_{S(t)^-}
\]  

(3.13)

which is the Stefan condition as described by Eqn (3.3).

3.2.2 The Neumann solution: an analytical result

The above described system corresponds to the classical formulation of the two-phase Stefan problem in a planar geometry, one of the few cases for which an analytical solution is available (Crank, 1987; Gupta, 2003).

The Neumann solution, as it is called, considers a finite or semi-infinite domain and solves the freezing problem for a one-dimensional geometry (a slab). It is a well
known similarity solution given by (Alexiades and Solomon, 1993):

\[ S(t) = 2\eta \sqrt{\alpha_s t} \]  

(3.14)
equation which provides the front position at each time step. The constant \( \eta \) is the solution of the following transcendental equation (Voller and Cross, 1980):

\[ e^{-\eta^2} \frac{\text{erf}(\eta)}{\text{erf}(\eta)} - \frac{k_l}{k_s \sqrt{\alpha_t}} \frac{(T(x,0) - T_f)e^{-\alpha_s \eta^2}}{(T_f - T(0,t)) \text{erfc} \left( \eta \sqrt{\frac{\alpha_s}{\alpha_t}} \right)} = \frac{\eta \Delta H \sqrt{\pi}}{C_p_s \left( T_f - T(0,t) \right)} \]  

(3.15)

which can be solved by employing a Newton iteration. The temperature distribution is given, piece-wisely, by:

\[
T(x, t) = \begin{cases} 
T_f - T(0, t) \frac{x}{2 \sqrt{\alpha_t t}} \text{erf} \left( \frac{x}{2 \sqrt{\alpha_s t}} \right), & x < S(t) \\
T_f, & x = S(t) \\
T(x, 0) - T_f \frac{x}{2 \sqrt{\alpha_t t}} \text{erfc} \left( \eta \sqrt{\frac{\alpha_s}{\alpha_t}} \right), & x > S(t)
\end{cases}
\]  

(3.16)

In Figure 3.2, both the evolution in time of the moving front, as well as the temperature history at \( x = 0.5 \)m, are showed. System data and parameters have been taken from (Voller and Cross, 1980).

![Figure 3.2: Analytical results for the freezing problem: a) moving front evolution in time b) temperature history at \( x = 0.5 \)m.](image-url)
3.3 Freezing problem: numerical solutions

The problem described in Section 3.2 will be numerically undertaken by employing three different methods:

- A *front-tracking* scheme, which makes use of an Arbitrary Lagrangian-Eulerian (ALE) algorithm (Kjellgren and Hyvarinen, 1998; Donea et al., 2004) to adapt the mesh to the front advance.

- A *front-fixing* method, which is based on a variable transformation to fix the computational space subdomains (Landau, 1950; Illingworth and Golosnoy, 2005).

- A *fixed-domain* method, the so called Enthalpy method, a technique which employs the enthalpy function $H(T)$ to account for the total energy of the system and so reformulating the problem for the whole spatial domain (Hunter, 1989; Nedjar, 2002).

For the sake of validation, the solutions obtained will be compared to the analytical results presented in Section 3.2.2. It must be noted that the analytical solution considers an semi-infinite domain. However, an approximation to a finite slab has been assumed for the numerical simulations. As justified in Alexiades and Solomon (1993), imposing an insulated boundary at $x = L$ is required for Eqn (3.7) to hold in the finite domain. Then, by means of Eqn (3.16) one can be obtained, either

![Figure 3.3: Distributed temperature at three different times obtained through the analytical solution for the freezing case. Results for $x \in [0, 8]$.](image-url)
3.3. Freezing problem: numerical solutions

numerically or graphically, a suitable length $L$. For the case here solved, it was found that with a length $L = 8$ the finite domain $\Omega$ can be considered to be semi-infinite, as depicted in Figure 3.3.

It is interesting also to comment that since the three methodologies selected allow their application in combination with the Finite Element method, this technique has been chosen as part of the numerical strategies followed to solve the benchmark proposed. In addition, standard spatial and time grids have been defined, being $\delta x = 0.1\ m$ and $\delta t = 1\ day$ the space and time steps, respectively.

3.3.1 A front-tracking approach

A numerical scheme combining the Finite Element Method (FEM) and an Arbitrary Lagrangian Eulerian (ALE) algorithm has been chosen from the front-tracking family of methods.

The ALE algorithm combines both motion descriptions, Lagragian and Eulerian, in order to establish a mapping between the so-called referential domain and the material and spatial domains, making use of subsequent refinements if the deformations of the spatial grid are significant (Donea et al., 2004).

An adaptive mesh and linear Lagrange P1 finite elements have been employed to discretised the one-dimensional spatial domain $\Omega = [0, L]$. The computation of the FE method, and the coupling with the ALE algorithm have been accomplished by means of the commercial software COMSOL Multiphysics (COMSOL AB, 2008), which provides an specific application mode for modelling moving boundary geometries.

The results obtained through the moving mesh have been spatially interpolated so both space and time grids coincide with the ones chosen as standard. Moreover, the analytical solution for $t = 1\ day$ has provided the initial conditions required to solve the problem.

Figure 3.4 shows the comparison between this front-tracking based solution and the corresponding Neumann one, with a close agreement between them.

Difficulties associated to front-tracking schemes

Although accurate, front-tracking methods also entail a list of drawbacks to be taken into account. The first of them is the computational burden related to remeshing the time-dependent spatial subdomains, as new front locations are tracked, specially when 2D or 3D geometries are considered. Another problem regarding multidimen-
Figure 3.4: Comparison between analytical (solid line) and front-tracking (marks) methods for the freezing problem: a) moving front evolution in time b) Temperature history at \( x = 0.5 \text{m} \).

Spatial geometries is related to the complex shapes that the moving front could adopt, becoming curvature a new factor to consider, as it is revealed by the expression of the Stefan condition for a two-dimensional spatial domain \( \Omega = \{(x, y) \in \mathbb{R}^2 | 0 \leq x \leq D, 0 \leq y \leq L, t > 0 \} \):

\[
\Delta H \rho_s \frac{\partial S}{\partial t}(x, t) = \left[ -k_l \frac{\partial T_l}{\partial y} \bigg|_{S(x,t)+} + k_s \frac{\partial T_s}{\partial y} \bigg|_{S(x,t)-} \right] \left[ \left( \frac{\partial S(x,t)}{\partial x} \right)^2 + 1 \right] \tag{3.17}
\]

where the front shape is parametrized by defining a function \( \gamma(x, y, t) = y - S(x, t) = 0 \), being \( y = S(x, t) \) the moving front position.

For the 3D case, with \( \Omega = \{(x, y, z) \in \mathbb{R}^3 | 0 \leq x \leq D, 0 \leq y \leq W, 0 \leq z \leq L, t > 0 \} \), the Stefan condition reads:

\[
\Delta H \rho_s \frac{\partial S}{\partial t}(x, y, t) = \left[ -k_l \frac{\partial T_l}{\partial z} \bigg|_{S(x,y,t)+} + k_s \frac{\partial T_s}{\partial z} \bigg|_{S(x,y,t)-} \right] \left[ \left( \frac{\partial S(x,y,t)}{\partial x} \right)^2 \right. \\
+ \left. \left( \frac{\partial S(x,y,t)}{\partial y} \right)^2 + 1 \right] \\
+ \left( \frac{\partial S(x,y,t)}{\partial z} \right)^2 \tag{3.18}
\]

after introducing the corresponding parametrisation \( \gamma(x, y, z, t) = z - S(x, y, t) = 0 \), where \( z = S(x, y, t) \) and \( \text{re}S(x, y, t) \) represents the surface defining the moving front.

The complete derivation of the multidimensional Stefan condition can be found in Appendix A.
In addition, the coexistence of the two phases at the initial time it is also needed for the Stefan condition to hold. In few cases, as in the example here presented, the analytical solutions can be helpful, but most of the times it will be required to find a suitable initial temperature distribution, with the corresponding front position which fulfils the system requirements, what is not always easy to do. However, there have been reported efforts on finding proper initialisation strategies for one-dimensional problems (Mitchell and Vynnycky, 2009).

![Figure 3.5: One-dimensional fixed domain resulting of applying the Landau transform.](image)

### 3.3.2 A front-fixing method: the Landau transform

By a suitable choice of space coordinates (Crank, 1987), the Landau transform (Landau, 1950; Illingworth and Golosnoy, 2005) enables a new spatial working domain where all the boundaries are fixed. For a two-phase problem like the one here considered, and aiming at immobilising the moving front while preserving the fixed nature of the edges, two new spatial coordinates (one for each region) are defined as:

\[
    z_s = \frac{x}{S(t)}; \quad z_l = \frac{x - S(t)}{L - S(t)}, \quad \forall x \in \Omega \tag{3.20}
\]

where \(z_s, z_l \in [0, 1]\). Consequently, the front will be kept motionless at \(z_s = 1, z_l = 0\) all along the computational time.

In this new framework, proper spatial and time operators are described in order to establish the mapping from the original time-varying domain to the new fixed one
(i.e. from $T_i(x,t)$ to $\bar{T}_i(z,\theta)$, being $i = s, l$ according to the corresponding phase):

\[
\frac{\partial T_s}{\partial x} = \frac{1}{S(t)} \frac{\partial T_s}{\partial z_s}; \quad \frac{\partial^2 T_s}{\partial x^2} = \frac{1}{(S(t))^2} \frac{\partial^2 T_s}{\partial z_s^2} \tag{3.21}
\]

\[
\frac{\partial T_s}{\partial t} = \frac{\partial T_s}{\partial \theta} \frac{\partial \theta}{\partial t} + \frac{\partial T_s}{\partial z_s} \frac{\partial z_s}{\partial t} = \frac{x}{(S(t))^2} \dot{S}(t) \frac{\partial T_s}{\partial z_s} \tag{3.22}
\]

\[
\frac{\partial T_l}{\partial x} = \frac{1}{L - S(t)} \frac{\partial T_l}{\partial z_l}; \quad \frac{\partial^2 T_l}{\partial x^2} = \frac{1}{(L - S(t))^2} \frac{\partial^2 T_l}{\partial z_l^2} \tag{3.23}
\]

\[
\frac{\partial T_l}{\partial t} = \frac{\partial T_l}{\partial \theta} \frac{\partial \theta}{\partial t} + \frac{\partial T_l}{\partial z_l} \frac{\partial z_l}{\partial t} = \frac{(L - x)}{(L - S(t))^2} \dot{S}(t) \frac{\partial T_l}{\partial z_l} \tag{3.24}
\]

with typically $\partial \theta / \partial t = 1$. The above relationships, once substituted in Eqns (3.1) and (3.2), lead to the following governing equations:

\[
\frac{\partial \bar{T}_s}{\partial t}(z_s, t) = w(t) \frac{z_s}{S(t)} \frac{\partial T_s}{\partial z_s}(z_s, t) + \frac{\alpha_s}{S(t)^2} \frac{\partial^2 T_s}{\partial z_s^2}(z_s, t) \tag{3.25}
\]

\[
\frac{\partial \bar{T}_l}{\partial t}(z_l, t) = w(t) \frac{1 - z_s}{(L - S(t))} \frac{\partial T_l}{\partial z_l}(z_l, t) + \frac{\alpha_l}{(L - S(t))^2} \frac{\partial^2 T_l}{\partial z_l^2}(z_l, t) \tag{3.26}
\]

since $\dot{S}(t) = w(t)$ and $L - x = (1 - z_l)/(L - S(t))$. Likewise, the boundary conditions will take the form:

\[
\frac{k_s}{S(t)} \frac{\partial \bar{T}_s}{\partial z_s}(1, \theta) - \frac{k_l}{L - S(t)} \frac{\partial \bar{T}_l}{\partial z_l}(0, \theta) = \rho_s \Delta H w(t) \tag{3.27}
\]

\[
\bar{T}_s(0, \theta) = \bar{T}_c; \quad \frac{k_l}{L - S(t)} \frac{\partial \bar{T}_l}{\partial z_l}(1, \theta) = 0 \tag{3.28}
\]

Due to the transformed spatial domain does not present time dependency, the FEM matrices have been computed by means of the matfem function, where linear Lagrange elements have been chosen. Once discretised, the resulting ODE system has been solved by making use of the Matlab solver ode15s. The outcomes obtained are shown in Figure 3.6.

**Difficulties associated to Landau transform technique**

Most of the inconveniences associated to the Landau transform are similar to those before listed when the front-tracking methods were analysed, for the Stefan condition is part of the problem formulation.

Accordingly, the problems related to curvature or irregular front shape in two and
3.3. Freezing problem: numerical solutions

Figure 3.6: Comparison between analytical (solid line) and Landau transform-based (marks) methods for the freezing problem: a) moving front evolution in time b) Temperature history at $x = 0.5m$.

three-dimensional geometries are characteristic of this method, too. As example, it follows the expression of the Stefan condition for a 2D case, which has been derived from Eqn (3.29):

$$\rho_s \Delta H w(x, \theta) = \left[ \frac{k_s}{S(t)} \frac{\partial \bar{T}_s}{\partial z_s}(1, \theta) - \frac{k_l}{L - S(x, \theta)} \frac{\partial \bar{T}_l}{\partial z_l}(0, \theta) \right] \left[ \left( \frac{\partial S(x, \theta)}{\partial x} \right)^2 + 1 \right]$$

(3.29)

The application of the Landau transform to a particular 2D domain can be found in Appendix B.

Besides, front-fixing approaches also require to initialise the problem after the moving boundary has already gone through a certain piece of material.

Although, the Landau transform offers advantages over the front-tracking approaches. Firstly, the computational burdens are lighten since there are no need of re-meshing to accommodate the spatial grid to the front motion, which could be relevant for real-time applications.

Furthermore, the transformation into a spatial domain with fixed boundaries enables the application of standard numerical methods for solving PDE systems (like those ones described in previous chapters), widening the range of options from which select the approach that better fits the problem features.
3.3.3 A fixed-domain technique: the Enthalpy method

The Enthalpy method is one of the most popular fixed-domain methods due its reported simplicity and flexibility for solving Stefan problems (Kutulay and Esen, 2004). It consists of introducing the enthalpy function, \( H(T) \), which accounts for the total energy of the system, into the problem formulation. This results into a new single governing equation, which applies for the whole spatial domain, and such that the Stefan condition is automatically satisfied across the moving front (Crank, 1987; Caldwell and Kwan, 2004) (proof can be found in Appendix C). This transformation is given by Fikiin (1996):

\[
H(T) = \int_{T_{ref}}^{T} \rho C_p \, dT
\]  

(3.30)

where \( T_{ref} \) is a suitable chosen reference so \( H(T_{ref}) = 0 \).

In order to deal with the existence of different thermal conductivities for each phase, the Kirchhoff transformation (Fikiin, 1996; Caldwell and Kwan, 2004) will be also employed. The new variable \( E(T) \) is defined as follows:

\[
E(T) = \int_{T_{ref}}^{T} k \, dT
\]  

(3.31)

Accordingly, \( E_{ref} \) is the reference value for the Kirchhoff’s function corresponding to \( T_{ref} \), which has been particularly defined as \( E_{ref} = E(T_{ref}) = 0 \).

Both Eqns (3.30) and (3.31) lead to the following piecewise functions for extensive variables enthalpy and Kirchhoff, respectively:

\[
H(T) = \begin{cases} 
H_{ref} + \rho_s C_{ps} (T_f - T_{ref}) + \rho_l C_{pl} (T - T_f) + \Delta H \rho_s & \text{if } T > T_f \\
\in [H_{ref}, H_{ref} \Delta H \rho_s] & \text{if } T = T_f \\
H_{ref} + \rho_s C_{ps} (T - T_{ref}) & \text{if } T < T_f 
\end{cases}
\]  

(3.32)

\[
E(T) = \begin{cases} 
E_{ref} + k_s (T_f - T_{ref}) + k_l (T - T_f) & \text{if } T > T_f \\
E_{ref} + k_s (T_f - T_{ref}) & \text{if } T = T_f \\
E_{ref} + k_s (T - T_f) & \text{if } T < T_f 
\end{cases}
\]  

Since the thermophysical properties are positive and bounded, and the temperature field is strictly monotonic, there exist a one-to-one mapping between dependent
3.3. Freezing problem: numerical solutions

and independent variables. Only dependences $H(T)$ and $H(E)$ exhibit a jump discontinuity at $T = T_f$ (Fikiin, 1996).

Differentiation in time at both sides of Eqn (3.30) and Eqn (3.31) and further application of the Leibniz rule to the RHS of both equations render to:

$$\frac{\partial H}{\partial t} = \rho C_p \frac{\partial T}{\partial t}; \quad \frac{\partial^2 E}{\partial x^2} = k \frac{\partial^2 T}{\partial x^2}$$

(3.33)

The substitution of these equivalences into Eqn (3.1) and (3.2) results into a unique governing equation for the whole domain:

$$\frac{\partial H}{\partial t} = \frac{\partial^2 E}{\partial x^2}, \quad x \in (0, L)$$

(3.34)

The complementary initial and boundary conditions are obtained by transforming the originally temperature-based conditions according to the piece-wise functions $H(T)$ and $E(T)$. Therefore, the extensive system formulation reads as follows:

$$\frac{\partial H}{\partial t} = \frac{\partial^2 E}{\partial x^2}, \quad x \in (0, L)$$

(3.35)

$$E(0, t) = E_c; \quad \frac{\partial E}{\partial x}(L, t) = 0$$

(3.36)

Finally, suitable enthalpy initial conditions close the system:

$$H(x, 0) = H(T(x, 0))$$

(3.37)
In order to compute the corresponding numerical solution, once again, the *matfem* function is called in order to obtain the FEM matrices. Integration in time of the discrete system has been performed by means of the implicit solver *ode15s*. The front position evolution has been computed by a post-processing stage through numerical interpolation of the temperature value corresponding to the phase change at each time step. Figure 3.8 shows the results obtained when using the standard grids. Note the evident deviations in the results, as well as the characteristic stepwise behaviour of the solutions when interpolation is chosen to locate the front (Crank, 1987). However, by increasing the number of nodes in the spatial grid, it is possible to achieve the accuracy required, as show in Figure 3.9.

**Problems and advantages of the Enthalpy-Kirchhoff approach**

As just pointed out, the mixed Enthalpy-Kirchhoff approach needs of finer spatial grids in order to achieve the same accuracy as other techniques presented in Sections 3.3.1 and 3.3.2, which constitutes the main disadvantage of this method.

On the other hand, there are some advantages rendered by the the implicit consideration of the Stefan condition into the enthalpy-based formulation of the problem. Since the transformed problem is defined on a fixed domain where the moving interface is not longer considered, no problems with complex shapes and curvature of the front can be encountered. Thus multidimensional problems in 2D or 3D geometries can be solved by employing standard numerical methods, such those ones introduced in Chapters 1 and 2. In this new framework, the front position can be considered...
3.4 Model reduction for moving boundary problems

Figure 3.9: Comparison between analytical (solid line) and Landau transform-based (dashed) methods for the freezing problem: a) moving front evolution in time b) Temperature history at $x = 0.5\text{m}$ when a spatial grid with 321 nodes is employed.

as a feature derived from the temperature distribution, for it can be computed in a post-processing stage. In addition, the bypass of the Stefan condition also simplifies the initialisation tasks.

It is also remarkable the suitability of the method to deal with mushy regions (i.e. phase change phenomena occurring in a given range of temperatures), as well as to solve problems involving temperature dependent properties ($k, \rho, C_p$), for which the combination of both transformations, enthalpy and Kirchhoff, eases the computation significantly (Crank, 1987; Hu and Argyropoulos, 1996).

3.4 Model reduction for moving boundary problems

The applicability of the reduced order methods (ROM) presented in Chapter 2 to problems exhibiting time-dependent spatial domains will be examined next.

Both model reduction techniques considered, LSD and POD, involve the projection of the PDE system onto the corresponding set of global basis functions. This projection requires integration over the spatial domain, step which constitutes the main challenge when dealing with time-varying domains.

As seen in previous sections, it is possible to reformulate the moving boundary problem into a domain with fixed boundaries, where projection could be realised in a straightforward manner. Either the Landau transformation or the Enthalpy method will provide a suitable framework for that purposes.

Model reduction in one-dimensional reaction-diffusion systems with one moving
boundary can be found in Armaou and Christofides (1999), Armaou and Christofides (2001a) and Armaou and Christofides (2001b). In this series of works, the PDE system is first represented on the domain with fixed boundaries provided by the Landau transform, and then the Proper Orthogonal Decomposition (POD) is applied in order to obtain a time-independent set of basis functions to be projected onto the transformed domain.

On the contrary, the adaptive mesh schemes will require to translate the grid time dependence into the set of basis functions, since now they will not only exhibit spatial but also temporal dependence. In some cases, this can be accomplished by giving an ALE-based numerical treatment to the POD basis (Ng et al., 2013; Stankiewicz et al., 2013).

One of the main novelties presented in this thesis deals with the application of model reduction techniques to the particular case of Stefan problems. This has been a topic poorly addressed in literature yet, probably because of the difficulties that can be found in handling the Stefan condition and its thermodynamic role. Accordingly with the model reduced schemes previously exposed, both Landau transformation and Enthalpy method will be combine with POD and LSD reduction techniques, respectively, in order to provide low-dimensional representations for the case study consisting of a phase-change problem.

In addition, there exist similarities between the Enthalpy-Kirchhoff transformed system and the travelling wave systems that could be exploited, specially regarding numerical resolution methods. In this framework, the template fitting technique (Rowley and Marsden, 2000) constitutes an interesting option to be explored which might bring advantages when considering model reduction. For the sake of illustration, this technique has been applied in Appendix D to a convection-diffusion system, and a first approach to the extensive formulation of the Stefan problems will be also presented, among other numerical schemes based on reduced models, in upcoming Part II.
Part II

Operational models for control applications
The previous chapters have introduced the modelling of distributed systems and techniques to obtain their low-dimensional representations. In the following ones, this background will be employed to derive operational models for those systems which constitutes the object of study of this thesis.

Those operational models are intended to provide not only accurate representations of the systems, but also computationally efficient ones, for they will be employed for real time purposes.

The general strategy in the obtaining of the operational models for the different systems presented starts with the development of a detailed mathematical description of the system dynamics based on first principles. Once the predictive capabilities of this first representation are ensured, it follows the employment of model reduction techniques. In this framework, both approaches, physical and mathematical will be followed. In this way, and based on the knowledge of the system, certain simplifications and assumptions must be adopted with the aim of collecting the sufficient features of the system needed to reproduce its behaviour with rigour. In addition, mathematical model reduction techniques will be further applied to reduce the dimensionality of the system, lessening computational times while preserving the accuracy of the representation.

Chapter 4 presents the operational model developed which describes the operation of a heat-exchanger reactor. The steps detailed above will be detailed, and in addition the optimal control of the reactor operation will be presented as example of real time application.

On the other hand, Chapters 5, 6 and 7 are focused on the application of the
proposed modelling scheme to moving boundary problems. Specifically they will be considered those problems exhibiting a moving front associated to a phase-change, namely Stefan problems. The development of a model based on the analysis of the time scales for freeze-drying processes will be presented in Chapter 5, while Chapter 6 is devoted to the optimal control of the process together with the development of new methodology to estimate on-line, and in a non-invasive way, the sample temperature distribution.

Additionally, in Chapter 7 the extensive formulation involving Enthalpy and Kirchhoff variables will be not only applied to the freeze-drying case study, but also to a generic phase change problem in order to introduce the application of model reduction techniques for Stefan problems. In this framework, a first approach to the application of the template-fitting technique to the extensive formulation of the Stefan problems will be also presented.
Introducción

Os capítulos anteriores introduciron a modelaxe de sistemas distribuídos e técnicas para obter as súas representacións de baixa dimensionalidade. A continuación, esta base será empregada na derivación modelos operacionais para os sistemas que constitúen o obxecto de estudo desta tese.

Estes modelos operacionais teñen a función de proporcionar non só representacións precisas dos sistemas, senón tamén computationalmente eficientes, xa que van ser empregados para aplicacións en tempo real.

A estratexia xeral na obtención dos modelos operacionais para os distintos sistemas presentados comeza co desenvolvemento dunha descripción matemática detalhada da dinámica do sistemas baseada en primeiros principios. Unha vez que queden aseguradas as capacidades predictivas desta primeira representación, o seguinte paso será o emprego de técnicas de redución de modelos. Neste marco, dous enfoques distintos, físico e matemático van ser combinados. Deste xeito, e fundamentadas nun coñecemento do sistema, algunhas simplificacións e suposicións deben ser adoptadas co fin de capturar as características suficientes do sistema necesarias para reproducir o seu comportamento con rigor. Ademais, tamén se aplicarán técnicas matemáticas de redución de modelos para rebaixar aínda máis a dimensionalidade do sistema, diminuíndo os tempos computacionais e conservando a precisión da representación.

O Capítulo 4 presenta o modelo operacional desenvolvido para describir o funcionamento dun reactor intercambiador de calor. Describiranse os pasos descritos anteriormente e, complementariamente, presentarase un problema de control óptimo como exemplo de aplicación en tempo real.

Por outra banda, os Capítulos 5, 6 e 7 están orientados á aplicación do esquema de
modelaxe proposto para problemas de fronteira móbil. En concreto, van ser abordados aqueles problemas caracterizados pola existencia dunha fronte en movemento asociada a un cambio de fase, é dicir, os denominados problemas de Stefan. No Capítulo 5 vaise presentar un modelo baseado na análise das escalas de tempo para procesos de liofilización, namentres que o Capítulo 6 está adicado ás aplicacións en tempo real do modelo operacional, isto é, ó control óptimo do proceso e ó desenvolvemento dunha nova metodoloxía para a estimación en-liña de forma non invasiva da distribución de temperatura do producto.

Finalmente, no Capítulo 7 vaise aplicar a transformación en variables extensivas (entalpía e variable de Kirchhoff) non só para o estudio do problema de liofilización, senón tamén a un problema de cambio de fase xenérico a fin de introducir a aplicación de técnicas de redución de modelos no ámbito dos problemas Stefan. Neste marco, presentarase tamén unha sínxelo exemplo que debe ser tomado como paso previo na aplicación da técnica do template-fitting para a formulación en variables extensivas dos problemas de Stefan.
4.1 Introduction

In this chapter, the mathematical description of a tubular reactor presented in Chapter 1 is recovered to be employed as the starting point in the procedure of obtaining a suitable model for optimisation purposes of the so-called Open Plate Reactor (OPR).

The OPR (Haugwitz, 2007) is a compact heat exchange reactor developed by Alfa Laval AB which combines intensified mixing with enhanced heat transfer capacity into one operation. These features suit perfectly for carrying out processes involving highly exothermic reactions, enabling the use of much more concentrated reactants and hence decreasing the efforts spent in subsequent separation stages.

The challenge inherent to the OPR relies on satisfying safety operation conditions during the start-up phase, since the exothermic nature of the reactions produced, and the difficulty in managing the reactor dynamics at such early times of process, can easily lead to dangerous high temperatures inside the reactor.

The modelling and control of the OPR have received particular attention in recent years. Experimental-based studies have been developed in order to characterise the dynamics of the reactor (Bouaifi et al., 2004; Andersson et al., 2004; Pratt et al., 2005). Model predictive control has been covered in Haugwitz et al. (2007) and Rodriguez et al. (2008), being the POD method for model reduction introduced in the latter one. In Haugwitz et al. (2009) dynamic optimisation has been used to obtain start-up trajectories and a feedback control system has been also presented, while in Lopez-Quiroga et al. (2010) further reduction of the system together with a comparison between the performance of the LSD versus POD methods has been reported.
Chapter 4

The mathematical model for the OPR can be derived from first principles by approximating its dynamical behaviour by a continuous tubular reactor. As a result, a strongly non-linear PDE system, involving temperature and species concentrations, is obtained. Due to the high dimensionality of the resulting model, the computation times needed in the numerical simulation when classical methods of solving PDE’s are employed, e.g. FDM or FEM, act as a deterrent against on-line applications.

In this chapter, both physical and mathematical reduction approaches, with strongest emphasis in the latter one, will be considered in order to derive an operational model suitable for control/optimisation tasks. The LSD and POD techniques will be employed and its performance analysed. The resulting reduced representation of the system will become the core of a real time optimisation (RTO) strategy for controlling the OPR operation subject to safety requirements.

4.2 Model description

The OPR design is based on plate heat exchangers, which separate the hot and cold fluids in alternate plates. The mixing of the reactants is performed in the so called service plates, while the cooling fluid flows through the utility ones. These latter ones are located at both top and bottom of each service plate, as depicted in Figure 4.1. Since the cross section of the horizontal channels in the OPR is too small compared with its length, an equivalent design consisting of a continuous plug reactor with cooling jacket, like the one represented in Figure 4.2, can be used (Haugwitz, 2007).

In this work, the following exothermic and irreversible reaction has been considered:

\[
2A + 4B \rightarrow C + D \tag{4.1}
\]

where \(A\) is the limiting reactant, being reactant \(B\) then in excess, and with \(C\) and \(D\) as the product species. The non-linear PDE-based model describing the dynamic behaviour of the tubular reactor-like system is obtained from the energy balances inside the reactor and inside the cooling jacket, together with the corresponding mass balances for each chemical specie involved in the reaction, from which follows a system of six partial differential equations.

After a preliminary analysis of the outlined model, further reduction can be still achieved by formulating the following assumptions:
4.2. Model description

Figure 4.1: Open Plate Reactor scheme. (1) Reactants are injected through the left upper reactor region, either in a unique point or in multiple ones. Temperature measurement points are represented by $Y_1$ and $Y_2$. (2) Schematic representation for the plates distribution: reaction plate in the middle surrounded by a couple of utility plates (Haugwitz, 2007).

Figure 4.2: Tubular reactor with cooling jacket scheme.

- **Only the energy balance inside the reactor will be considered.**

  Numerical simulation has shown that the evolution of the cooling temperature $T_c$ is properly defined by a constant value, therefore the energy balance for the cooling jacket can be neglected without affecting the reactor dynamics. However, when $T_c$ were considered as a control variable, it will be supposed to be a time dependent function, i.e. $T_c(t)$.

- **Product mass balances will be omitted.**

  From the stoichiometric relationships established in Eqn (4.1), the knowledge of the reactants concentrations leads to the immediate obtaining of the corresponding products concentrations. Thus, only reactants mass balances will be considered.

These physical simplifications, which rely on the insights of the system behaviour,
lead to a new system consisting only of three PDEs, which means halving the initial number of equations. The resulting dynamic model for the Open Plate Reactor reads as follows:

\[
\frac{\partial T_r}{\partial t}(z,t) = \alpha \frac{\partial^2 T_r}{\partial z^2}(z,t) - v_r \frac{\partial T_r}{\partial z}(z,t) + \frac{4h}{D_r \rho C_p} (T_c - T_r(z,t)) + \frac{\Delta H}{\rho C_p} r(C_A, C_B, T_r), \quad \forall z \in (0, L)
\]  

(4.2)

\[
\frac{\partial C_A}{\partial t}(z,t) = D_m \frac{\partial^2 C_A}{\partial z^2}(z,t) - v_r \frac{\partial C_A}{\partial z}(z,t) - 2r(C_A, C_B, T_r), \quad \forall z \in (0, L)
\]  

(4.3)

\[
\frac{\partial C_B}{\partial t}(z,t) = D_m \frac{\partial^2 C_B}{\partial z^2}(z,t) - v_r \frac{\partial C_B}{\partial z}(z,t) - 4r(C_A, C_B, T_r), \quad \forall z \in (0, L)
\]  

(4.4)

where \( z \) is the position in the reactor, \( T_r \) and \( T_c \) are the temperatures in the reactor and in the cooling jacket, respectively, whereas \( C_A \) and \( C_B \) note the reactants concentrations.

From the experiments in Bouaifi et al. (2004) and in Andersson et al. (2004), the Arrhenius law holds as a valid approximation for the reaction kinetics (Haugwitz, 2007). Thus, the reaction term \( r(C_A, C_B, T_r) \) is given by:

\[
r(C_A, C_B, T_r) = k_0 C_A C_B \exp \left( \frac{-E_a}{RT_r} \right)
\]  

(4.5)

Boundary conditions for the reactor inlet are defined in the form of a non-homogeneous Robin condition:

\[
\frac{\partial T_r}{\partial z}(0, t) = \frac{v_r}{\alpha} (T_r(0, t) - T_m) ; \quad \frac{\partial C_J}{\partial z}(0, t) = \frac{v_r}{D_m} (C_J(0, t) - C_{Jm})
\]  

(4.6)

while for the outlet reactor, Neumann homogeneous conditions have been imposed:

\[
\frac{\partial T_r}{\partial z}(L, t) = 0 ; \quad \frac{\partial C_J}{\partial z}(L, t) = 0
\]  

(4.7)

with \( J = \{A, B\} \) noting the reactant species. Finally, the model is closed by the corresponding initial conditions:

\[
T_r(z, 0) = T_0, \quad C_J(z, 0) = C_{J0}
\]  

(4.8)
### 4.2 Model description

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$</td>
<td>Reacting mixture density $1000 \text{ m}^3/\text{kg}$</td>
</tr>
<tr>
<td>$C_p$</td>
<td>Reacting mixture heat capacity $4180 \text{ J/kgK}$</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Reacting mixture thermal diffusivity $0.037 \text{ m}^2/\text{s}$</td>
</tr>
<tr>
<td>$h$</td>
<td>Heat transfer coefficient $3000 \text{ W/m}^2\text{K}$</td>
</tr>
<tr>
<td>$E_a$</td>
<td>Activation energy $76000 \text{ J/mol}$</td>
</tr>
<tr>
<td>$D_m$</td>
<td>Mass diffusion coefficient $0.037 \text{ m}^2/\text{s}$</td>
</tr>
<tr>
<td>$D_r$</td>
<td>Reactor diameter $0.0081 \text{ m}$</td>
</tr>
<tr>
<td>$\Delta H_r$</td>
<td>Reaction heat $1.17e^6 \text{ J/mol}$</td>
</tr>
<tr>
<td>$v_r$</td>
<td>Flow velocity $0.27 \text{ m}^2$</td>
</tr>
<tr>
<td>$L$</td>
<td>Reactor length $8.1 \text{ m}$</td>
</tr>
<tr>
<td>$C_{A_{nom}}$</td>
<td>Nominal inlet concentration reactant A $2500 \text{ mol/m}^3$</td>
</tr>
<tr>
<td>$C_{B_{nom}}$</td>
<td>Nominal inlet concentration reactant B $19135 \text{ mol/m}^3$</td>
</tr>
<tr>
<td>$T_{B_{in}}$</td>
<td>Inlet temperature reactant B $20^\circ \text{C}$</td>
</tr>
<tr>
<td>$q_{A_{in}}$</td>
<td>Reactant A feed flow $1.11e^{-5} \text{ m}^3/\text{s}$</td>
</tr>
<tr>
<td>$q_{B_{in}}$</td>
<td>Reactant B feed flow $2.78e^{-6} \text{ m}^3/\text{s}$</td>
</tr>
</tbody>
</table>

Table 4.1: Parameters for the OPR model.

### 4.2.1 Model validation

In order to validate the model proposed, one of the motivating examples presented in Haugwitz (2007) has been chosen as reference to compare with. It corresponds to the OPR start-up operation carried out under prescribed variations of the reactant A feeding temperature, $T_{A_{in}}$, and the percentage of reactant B injected. Such time profiles are depicted in Figure 4.3. Moreover, the single injection point for the specie B considered is located at the reactor inlet. To maintain stoichiometric conditions, the following relation will be considered:

$$q_{A_{in}} C_{A_{in}} = 0.5 q_{B_{in}} C_{B_{in}} \quad (4.9)$$

Initially, the reactor has been preheated to $20^\circ \text{C}$ and filled with reactant A. The cooling temperature has been set to $T_c = 20^\circ \text{C}$. The model presented in Eqn (4.2)-(4.8) has been solved by employing the Finite Element Method (FEM) according to the numerical procedure explained in Section 1.3.

The spatial domain has been discretised by employing a homogeneous grid consisting of 120 Lagrange P1 elements. In addition, the Matlab solver $ode15s$ has been employed to perform the time integration for $t = [0 : 1 : 200]$ seconds.

Results obtained closely reproduced the dynamic behaviour described in Haugwitz (2007). As can be seen in Figures 4.3 and 4.4, despite the continuous injection of reactant B, the reaction does not start until the feeding temperature $T_{A_{in}}$ is increased.
When $T_{A_{in}}$ reaches its maximum around $t = 100 \text{ s}$, the reactor temperature at the injection point rises fast, and the reaction then becomes self-accelerating.

Figure 4.4: Time profile corresponding to $T_r$ at the reactant B injection point for the OPR model validation.

4.3 Dimensionless model

In order to both ease the analysis and control of the OPR dynamic model, and to lessen the amount of parameters while achieving a well-conditioned system for numerical simulation, the model described by Eqns (4.2)-(4.8) is transformed into a
4.3. Dimensionless model

dimensionless one by considering the following changes in the independent variables:

\[ \xi = \frac{z}{L}; \quad \tau = \frac{v_r t}{L} \]  \hspace{1cm} (4.10)

where the length reactor, \( L \), and the flux velocity, \( v_r \), are the reference parameters chosen for the definition of the dimensionless space and time, \( \xi \) and \( \tau \), respectively.

The reactant concentrations, together with the reactor temperature, as well as the cooling temperature, have been also transformed according to expressions:

\[ x_A(\xi, \tau) = \frac{C_A - C_{A,\text{ref}}}{C_{A,\text{ref}}}; \quad x_B(\xi, \tau) = \frac{C_B - C_{B,\text{ref}}}{C_{B,\text{ref}}} \]
\[ y(\xi, \tau) = \frac{T_r - T_{\text{ref}}}{T_{\text{ref}}}; \quad y_c(\xi, \tau) = \frac{T_c - T_{\text{ref}}}{T_{\text{ref}}} \]  \hspace{1cm} (4.11)

Then, the dimensionless model equations can be written as:

\[ \frac{\partial y}{\partial \tau}(\xi, \tau) = \frac{1}{P_{e_1}} \frac{\partial^2 y}{\partial \xi^2}(\xi, \tau) - \frac{\partial y}{\partial \xi}(\xi, \tau) + W(y_c - y(\xi, \tau)) + \right) + D a_1 f(x_A, x_B, y), \ \forall \xi \in (0, 1) \]  \hspace{1cm} (4.12)

\[ \frac{\partial x_A}{\partial \tau}(\xi, \tau) = \frac{1}{P_{e_2}} \frac{\partial^2 x_A}{\partial \xi^2}(\xi, \tau) - \frac{\partial x_A}{\partial \xi}(\xi, \tau) - 2 D a_2 f(x_A, x_B, y), \ \forall \xi \in (0, 1) \]  \hspace{1cm} (4.13)

\[ \frac{\partial x_B}{\partial \tau}(\xi, \tau) = \frac{1}{P_{e_2}} \frac{\partial^2 x_B}{\partial \xi^2}(\xi, \tau) - \frac{\partial x_B}{\partial \xi}(\xi, \tau) - 4 D a_2 f(x_A, x_B, y), \ \forall \xi \in (0, 1) \]  \hspace{1cm} (4.14)

The non-linear term \( f(x_A, x_B, y) \) is defined as:

\[ f(x_A, x_B, y) = (1 + x_A)(1 + x_B)\exp\left(\frac{E_a}{RT_{\text{in}}} \left(\frac{y}{1 + y}\right)\right) \]  \hspace{1cm} (4.15)

and it plays the role of the reaction term. It follows from considering a reference rate of the form:

\[ r_{\text{ref}} = k_0 C_{A,n} C_{B,n} \exp\left(-\frac{E_a}{RT_{\text{in}}}\right) \]  \hspace{1cm} (4.16)

After the dimensionless transformation, the boundary conditions can be expressed as:

\[ \frac{\partial y}{\partial \xi}(0, \tau) = P_{e_1} (y(0, \tau) - y_m); \quad \frac{\partial x_J}{\partial \xi}(0, \tau) = P_{e_2} (x_J(0, \tau) - x_{J,m}) \]  \hspace{1cm} (4.17)
\[ \frac{\partial y}{\partial \xi}(1, \tau) = 0; \quad \frac{\partial x_J}{\partial \xi}(1, \tau) = 0 \]  

(4.18)

The model is completed by defining the initial conditions:

\[ y(\xi, 0) = y_0, \quad x_J(\xi, 0) = x_{J0}, \quad 0 \leq \xi \leq 1 \]  

(4.19)

<table>
<thead>
<tr>
<th>( P_e_1 )</th>
<th>Péclét number for the heat diffusive mechanism</th>
</tr>
</thead>
<tbody>
<tr>
<td>( P_e_2 )</td>
<td>Péclét number for the mass diffusive mechanism</td>
</tr>
<tr>
<td>( Da_1 )</td>
<td>Damköhler number for the heat convective mechanism</td>
</tr>
<tr>
<td>( Da_2 )</td>
<td>Damköhler number for the mass convective mechanism</td>
</tr>
<tr>
<td>( W )</td>
<td>Dimensionless group of parameters</td>
</tr>
</tbody>
</table>

Table 4.2: Dimensionless numbers for the OPR system.

4.3.1 Multi-injection system

As previously described in 4.2, OPR design enables multiple injection points. Aiming at preserving this feature for optimisation purposes, a piece-wise system configuration can be defined as the sum of \( n \) reactors (Álvarez, 2010) of length \( L_p \), each one of them described by the dimensionless model above presented in Eqns (4.12)-(4.19):

\[ \sum_{p=1}^{n} L_p = L; \quad p = 1, 2, ..., n \]  

(4.20)

where \( n \) accounts for the number of injections. This approach is illustrated in Figure 4.5. Accordingly, a geometrical factor \( a_p \), is also defined as:

\[ a_p = L_p/L; \quad \sum_{p=1}^{n} a_p = 1; \]  

(4.21)

In order to define each reactor section, a correction which consists of multiplying the dimensionless numbers in Table 4.2 by the factor \( a_p \), must be carried out.

4.3.2 System validation

The dimensionless reactor model with two injection points has been employed for numerical simulation of the OPR start-up operation. In order to validate the proposed model, the results obtained has been compared with previous works available
4.3. Dimensionless model

Figure 4.5: Multi-injection system scheme.

in literature. For the sake of clarity in the comparison, the physical variables have been employed to report the outcomes.

Four input variables have been taken into account: the amount of reactant B injected at both points, $b_1$ and $b_2$; the cooling temperature, $y_c$, and the reactant A feeding temperature, $y_{A_{in}}$. Besides, two different sets of values for those variables have been considered.

Figure 4.6: Steady-state temperature evolution along the OPR (solid) and conversion (dash) when the reactant B feeding is equally divided among two injection points.

Firstly, constant values for the four inputs have been taken from Haugwitz et al. (2007). The first injection point has been located at the reactor inlet, while the second one has been placed at half the reactor length. Each reactor section has been spatially discretised by employing the same grid already described in Section 4.2.1. Again, the Matlab solver ode15s has been employed for time integration in the corresponding time interval $t = [0 : 0.4 : 100]$ seconds. Although a good agreement is observed between both works, it must be mentioned that while the operational model here proposed assumes a constant value for the cooling temperature, in Haugwitz...
| Proposed model (Haugwitz et al., 2007) | 90.40 | 88.50 | 97.80 |
| Max 1 (K) | Max 2 (K) | Conversion (%) |

Table 4.3: OPR comparison results: proposed model versus literature, when $E_a = 68200 \text{ J/mol}$ with feeding reactant concentrations $C_{A_{in}} = 1470 \text{ (mol/m}^3\text{)}$ $C_{B_{in}} = 11256 \text{ (mol/m}^3\text{)}$ (Haugwitz et al., 2007)

et al. (2007) this variable is considered to evolve according the PDE resulting from the cooling jacket energy balance. Therefore, the final time value in that distributed profile has been set as the constant value for $T_c$ to be employed for numerical solving. As consequence, differences in the maxima reached by the reactor temperature around

![Fig 4.7](image-url)

Figure 4.7: Time profiles corresponding to the input variables for the second OPR validation case. Adapted from (Haugwitz et al., 2009): a) $b_1$, b) $b_2$, c) $T_c$ and d) $T_{A_{in}}$

the injections points, as well as regarding the conversion figures, are reported (see Table 4.3).

The second case considers a dynamic behaviour for the inputs. The corresponding time profiles have been adapted from Haugwitz et al. (2009), and they are shown in Figure 4.7. According to this reference work, when this set of controls is employed
4.4 Reduced order models

the reactant B do not overtake certain concentration values at injections points, and reactor temperature is always kept below the safety value.

However, the results obtained when the reported input profiles were employed strongly disagreed from those presented in Haugwitz et al. (2009). As seen in Figure 4.8, the restrictions regarding $C_B$ are largely violated. Moreover, and even though the reactor temperatures obtained are lower than the maximum allowed, they followed totally different trajectories.

![Figure 4.8: Evolution in time of the reactant B concentration at both injection points: a) first point, b) second point obtained by means of the proposed operational model when employing the inputs depicted in Figure 4.7. Red lines represent the limit values defined in Haugwitz et al. (2009).](image)

Consequently, the model had been gone through a complete checking, but no cause has been found to such mismatch. Nevertheless, and despite these adverse findings, it has been considered that the model reasonably reproduces the expected behaviour of the reactor. Therefore, the model will be employed for further applications.

4.4 Reduced order models

Even after having accomplished a severe physical reduction of the model, the system of ODE’s to be solved for a single reactor section amounts to 363 equations, one for each degree of freedom, which entails a significant computational effort, specially when real-time tasks are being carried out. This calls for the employment of model reduction-based methods of solving PDE’s systems.

Both techniques presented in Chapter 2, the Laplacian Spectral Decomposition and the Proper Orthogonal Decomposition, will be applied to the piece-master reactor
described by Eqns (4.12)-(4.19), since any multi-injection configuration can be derived from this one.

Accordingly, a single-injection dimensionless system will be considered. Through this point, the 80% of the reactant B feeding will be entered into the system. As in previous examples, the reactor is assumed to be initially full of reactant A and preheated to 20°C. In this Section, and for the sake of simplicity when dealing with the boundary conditions, the inlet concentrations \( C_{A_{in}}, C_{B_{in}} \) and temperature \( T_{in} \) have been chosen as the reference values \( C_{A_{ref}}, C_{B_{ref}} \) and \( T_{ref} \), respectively. This transforms the non-homogeneous boundary conditions defined in Eqn (4.17) into homogeneous ones, which together with Eqn (4.18) enables the application of the LSD method.

Based on the results obtained, the most efficient technique will be chosen as the core of the dynamic optimisation problem formulated in Section 4.5.

### 4.4.1 LSD computation

The method demands to solve an eigenvalue problem of the form shown in Section 2.4, which in a discretised matrix fashion, for the OPR, can be read as follows:

\[
\mathcal{M} \mathcal{M}^{-1} \left( \frac{1}{Pe} \mathcal{D} \mathcal{M} + \mathcal{B} \mathcal{M} \right) \Phi_i = \lambda_i \Phi_i, \quad i = 1, ..., neig
\]

(4.22)

where \( \lambda_i \) are the discrete eigenvalues, sorted by increasing value, \( \Phi_i \) are the discrete eigenfunctions and \( neig \) is the number of them to be considered. This problem has been solved through the \texttt{matlsd} function within the MatMOL toolbox.

It is important to note that Eqn (4.22) involves a generic Péclet number, \( Pe \), since for the current case study, and due to the thermophysical and geometric parameters of the system, the Péclet number for the heat transport, \( Pe_1 \), equals the Péclet number for the mass transport, \( Pe_2 \).

Right after, the equation system with boundary conditions is projected onto the computed set of eigenfunctions leading to a new ODE system to be solved:

\[
\frac{d m_y}{dt} = A m_y + W (y_c - m_y) + D a_1 \mathcal{F}
\]

(4.23)

\[
\frac{d m_{x_A}}{dt} = A m_{x_A} - 2 D a_2 \mathcal{F}
\]

(4.24)

\[
\frac{d m_{x_B}}{dt} = A m_{x_B} - 4 D a_2 \mathcal{F}
\]

(4.25)
4.4. Reduced order models

being:

\[ A = - \Phi^T \left( \frac{1}{P_e} D M + B M + C M \right) \Phi, \quad \Phi = [\Phi_1 | \ldots | \Phi_{neig}] \]

\[ \mathcal{F} = - \Phi^T M M F(X_A, X_B, \mathcal{Y}) \]

\[ \mathcal{Y}_c = - \Phi^T M M \mathcal{Y}_c \]

where \( X_A, X_B, \mathcal{Y} \) represent the discrete state variables and \( F(X_A, X_B, \mathcal{Y}) \) accounts for the discrete non-linear term. The following initial conditions complete the system:

\[ m_y(0) = \Phi^T M M \mathcal{Y}(0); \]

\[ m_{x_A}(0) = \Phi^T M M X_A(0); \]

\[ m_{x_B}(0) = \Phi^T M M X_B(0); \]

Results obtained are shown in Figure 4.9, where the lack of accuracy of the LSD in capture the dynamics of the reaction is clearly revealed, even for small reductions.

![Figure 4.9: Comparison in terms of the temperature evolution at the injection point in the OPR system between the FEM full model (solid) and two different LSD-based low-order representations: neig = 101 (dot) and neig = 81 (dash).](image)

**Role of the Péclet number**

As already mentioned, one of the dimensionless parameters of the dynamic model is the Péclet number, \( P_e \), which appears both in the energy as well as in the mass dimensionless balances. The Péclet number associated to the heat diffusive mechanism, \( P_e_1 \), depends on the reactor length, \( L \), flux velocity, \( v_r \), density, \( \rho \), heat capacity, \( C_p \),
and diffusivity coefficient, $\alpha$. On the other hand, the one related to the mass diffusion, the $Pe_2$, is defined by $L$, $v_r$ and $D_m$, as can be seen in Table 4.2. Thus, each single reactor is characterized by a different couple of Péclet numbers.

The Péclet number plays an important role on system reduction since, according to Eqn (4.22), the eigenvalues and eigenfunctions are determined by the diffusion coefficient $(1/Pe)$. 

$$
\begin{align*}
\text{Figure 4.10: Comparison between } \lambda_i \text{ obtained from Eqn (4.22) for OPR systems with } Pe = 7 \text{ and } Pe = 59.
\end{align*}
$$

From the analysis of the eigenvalues computed for the OPR with Péclets numbers defined in Table 4.2, where $Pe = 59$, it can be seen that all the $\lambda_i$ are in the same order of magnitude, which complicates the separation between fast and slow dynamics, so compromising the effective reduction of the system.

In order to illustrate this drawback, an analogous system characterized by a low $Pe$ number ($Pe = 7$) has been also analysed. In this case, there is clearly a spectral gap (Shvartsman and Kevrekidis, 1998; Armaou et al., 2008) as shown in Figure 4.10, where the eigenvalues corresponding to both systems are compared.

According to this, the LSD method will be more efficient for obtaining a model reduction when dealing with low Péclet values (i.e. reaction systems with an important diffusion mechanism), which is not the case of the system being studied.

### 4.4.2 POD computation

When the POD method is chosen as model reduction tool, the first step to take is to obtain a suitable set of snapshots which represents the dynamics of the system, also
4.4. Reduced order models

under perturbations. Due to the nature of the OPR behaviour, it has been necessary
to gather more information about the times the chemical reaction takes place at, that
is, where the change in the dynamics is stronger. Using the FEM-solved model, which
will be referred to as "full system", a set of numerical simulations has been designed
to collect the ensemble of data. The time intervals of solution have been refined
around the reaction times so to better capture the information related. Moreover,
values for the cooling temperature have been considered: $T_c \pm 5^\circ C, T_c \pm 10^\circ C$.

Once the collection of snapshots was available, this database was employed to
arrive at a set of empirical eigenfunctions by means of the matpod function within the
MatMOL toolbox. The number of eigenfunctions $\Phi_i$ was determined by the amount
of energy enclosed by the eigenvalues $\lambda_i$, figure which is one of the required inputs
of the matpod function, together with the snapshots and the FEM mass matrices. A
more detailed description of this function can be obtained at www.matmol.org.

![Figure 4.11: Comparison between the full temperature evolution in time at the injection point (solid) and the POD results (marks) for neig=31 in the OPR system.](image)

Projection of the PDE-based system defined by Eqn (4.12)-(4.19) onto the computed basis functions lead to an ODE system analogous in form to the one already given by Eqns(4.23)-(4.26). Although, it must be noted that, for the POD implementation, different basis functions were obtained for each field.

The temperature profile in time at the injection point for both full and reduced
model is presented in Figure 4.11, where it can be seen the good agreement between
them. As shown in, a reduction in the degrees of freedom of around a 75% for
each state has been achieved, while maintaining the accuracy with respect to the full
4.5 Dynamic optimisation

In this section, an optimal start-up strategy for the Open Plate Reactor (OPR) will be sought by means of dynamic optimisation. The main challenge in this regard deals with the establishment of safety operation conditions, since the exothermic nature of the reaction considered, and the difficulty in managing the reactor dynamics at such early times of process, can easily lead to dangerous high temperatures inside the reactor.

Consequently, the objective will be to find the set of time dependent control variables which ensure the transition from the initial point of the operation, where the reactor is cold and no reactant B is fed, to an optimal operating point with maximum reactant conversion (Haugwitz et al., 2009) while keeping the temperature inside the reactor within the security margins defined for the process.

To such purpose, the reactor configuration consisting of two injection points for reactant B is considered again. The fraction of reactant B introduced into the unit through each one of them will constitute the first control variables, which will be noted as \( b_1 \in [0, 1] \) and \( b_2 \in [0, 1] \), respectively. Note that the stoichiometric conditions for a two-injection points system will be guaranteed only if \( b_1 + b_2 = 1 \). This will be the desired scenario to operate with maximum reactant conversion.

Two more control variables will be defined (Haugwitz et al., 2009): the reactant A feeding temperature, \( T_{A_{in}} \), and the cooling temperature, \( T_c \). Therefore, \( m = 4 \) will be the final number of manipulated variables.

In order to lighten as much as possible the associated computational burdens, the dimensionless POD-based operational model previously presented will be employed to define the process dynamics, which will be further referred to as \( f (\dot{z}, z, v, p, t) = 0 \).

The proposed dynamic optimisation problem will be transformed into a Non-Linear Programming (NLP) problem by means of a direct method: the Control Vector Parametrisation approach (Vassiliadis, 1993; Vassiliadis et al., 1994). This method discretises and approximates the control variables through low order polynomials.

To that purpose, the selected operation time horizon is discretised by employing \( \beta \) points, so divided into \( \beta - 1 \) time intervals and, the control variables in \( v \) are approximated by piece-wise linear functions in each interval. The combined use of ROM-based descriptions of process dynamics together with a CVP method has been
4.5. Dynamic optimisation

successfully employed before in Balsa-Canto et al. (2004b).

Among the available optimisation algorithms to solve NLP, the eSS-SSm Enhanced Scatter Search solver (Egea et al., 2009) has been selected. This method, recently developed for solving non-linear dynamic optimisation problems, shares some features of the scatter search meta-heuristic. This algorithm is simpler and quite effective in helping to overcome typical difficulties of non-linear dynamic systems optimisation such as noise, flat areas, non-smoothness, and/or discontinuities. It provides a good balance of robustness and efficiency between the global and local search, outperforming other state-of-the-art methods.

4.5.1 Results

As aforementioned, the objective is to ensure a safe start-up operation for the Open Plate Reactor. In order to illustrate the challenge of controlling the temperature inside the reactor while trying to achieve the maxima reactant conversions, a first optimisation problem without restrictions has been solved. This scenario, which allows any temperature value inside the reactor, can be mathematically described as follows:

\[
\min_u J
\]

subject to:

\[
f(\dot{z}, z, u, p, t) = 0
\]

\[
0 \leq b_1 \leq 0.7 \quad 0 \leq b_2 \leq 0.7
\]

\[
\frac{20 - T_{ref}}{T_{ref}} \leq y_c \leq \frac{80 - T_{ref}}{T_{ref}}
\]

\[
\frac{30 - T_{ref}}{T_{ref}} \leq y_{Ain} \leq \frac{80 - T_{ref}}{T_{ref}}
\]

\[
\frac{30 - T_{ref}}{T_{ref}} \leq y_{Ain} \leq \frac{2 - T_{ref}}{T_{ref}}, \quad \frac{20 - T_{ref}}{T_{ref}} \leq y_c \leq \frac{2 - T_{ref}}{T_{ref}}
\]

where \( z \in \mathbb{R}^\alpha \) are the states, \( \dot{z} \) are their derivatives, \( v \in \mathbb{R}^{mu} \) is the control vector that includes \( b_1, b_2, y_c \) and \( y_{Ain} \), and \( p \in \mathbb{R}^{mp} \) are a given set of parameters.
The objective function to be minimised is defined as follows:

\[
J = \int_0^{\tau_f} \left[ \omega_1 x_A^2(N, \tau) + \omega_B x_B^2(N, \tau) + \omega_{b_1} b_1^2(\tau) + \omega_{b_2} b_2^2(\tau) \right] d\tau + \\
+ \omega_T \left( \sum_{k=1}^{\beta} \dot{y}_c^2(\xi, \tau) + \dot{y}_{A_{in}}^2(\xi, \tau) \right)
\]  \hspace{1cm} (4.33)

where the weights \( \omega \) are chosen such that the consumption of the reactants is driven as quickly as possible, thus enhancing conversion. The restrictions defined in Eqn (4.32) are taken into account as path constraints and introduced as a penalty in the second term of Eqn (4.33). Such requirements regarding the temperature derivatives are intended to smooth the corresponding manipulated profiles. The upper and lower bounds for the considered controls have been taken from Haugwitz et al. (2009). Note that such bounds, as well as the variables, have been conveniently transformed into their dimensionless counterparts.

Figure 4.12: Time profiles obtained for the control variables in the OPR operational model proposed when no constraints are imposed: a) \( b_1 \), b) \( b_2 \), c) \( T_c \) and d) \( T_{A_{in}} \).
4.5. Dynamic optimisation

In addition, it must be mentioned that the control variables have been approximated by employing 50 piecewise linear functions. This, together with the 45 POD’s employed per reaction section, results into a NLP problem with 2250 decision variables, which represents a reduction up to the 60% in the number of parameters to be optimised with respect to the FEM approach. This dimensional reduction has been also translated to the computational times, which in this way have been noticeably decreased while the accuracy of the results has been maintained. The optimal profiles for the control variables obtained for this first case are shown in Figure 4.12. These set of inputs leads to a complete conversion of the reactant A. However, an excess of reactant B has been observed at the reactor outlet. In addition, the maximum temperature reached inside the unit clearly overpasses the safety upper bound described by literature, as can be seen in Figure 4.13.

The formulation of second optimisation problem meets the need for controlling the reactor temperature by adding constraints on the state variables. The first of them is related to the safety temperature inside the reactor $y_{saef}$, which is the maximum value allowed for the reactor temperature. This requirement can be described as follows:

$$y(\xi, \tau) \leq y_{saef} = \frac{180 - \frac{1}{T_{ref}}}{T_{ref}}$$  (4.34)

Moreover, and following the strategy presented in Haugwitz et al. (2009), the concentration of reactant B is also subject to the following restrictions, which will
apply only at each one of the injection points:

\[ x_{B1}^{r1}(1, \tau) \leq \frac{1500 - C_{B_{ref}}}{C_{B_{ref}}} \], \quad x_{B2}^{r2}(1, \tau) \leq \frac{2500 - C_{B_{ref}}}{C_{B_{ref}}} \] \hspace{1cm} (4.35)

where the subscripts \( r1 \) and \( r2 \) are employed to distinguish between the first and the second reactor piece.

The figures for these constraints have been taken from the results obtained in Section 4.3.2, when the time dependent inputs reported in Haugwitz et al. (2009) were implemented. This is, the validation case in disagreement with literature. Besides, it must be taken into account that the control variables for this second scenario have been approximated by constant piecewise functions (steps).

![Figure 4.14: Optimal profiles corresponding to the results of the dynamic optimisation problem with constraints: a) \( b_1 \), b) \( b_2 \), c) \( T_c \) and d) \( T_{A_{in}} \)](image)

Then, the dynamic optimisation problem to be solved can be described by Eqns (4.27)-(4.35) together with the constraints defined by Eqn (4.34) and Eqn (4.31). Both contribute to the objective function if any of the limits is overpassed. The functional to be minimised is next defined:

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4.6 Conclusions

\[ J = \omega_1 x_A(N, \tau_{final}) + \int_0^{\tau_f} \left[ \omega_b (b_1^2(\tau) + b_2^2(\tau)) + \right. \\
+ \omega_y (y(\xi, \tau) - y_{safe}) + \omega_{r1} (x_{B1}(1, \tau) - x_{B1}^{r1}) + \\
+ \omega_{r2} (x_{B2}^{r2}(1, \tau) - x_{B2}^{r2}) \right] d\tau \]

being \( \omega_1 = 1000 \), with the other weights equal to the unity.

In this case, 50 steps have been employed to approximate the control variables, while the number of POD’s in the ROM were again 45, being the optimal profiles attained for each one of the controls shown in Figure 4.14. The convergence curve corresponding to this optimisation scenario is shown in Figure 4.15.

![Convergence of the objective function for the dynamic optimisation problem with constraints.](image)

Figure 4.15: Convergence of the objective function for the dynamic optimisation problem with constraints.

4.6 Conclusions

An operational model for the Open Plate Reactor has been derived and presented. Its performance has been validated by employing existing works in literature. In general, the model adequately reproduces the expected system behaviour. However, divergences with respect to literature have been found and reported.

Besides, two different model reduction techniques have been applied in order to
find the low-order representation of the OPR system: the Laplacian Spectral Decomposition (LSD) and the Proper Orthogonal Decomposition (POD). It has been found that the reduction power of the LSD method is strongly determined by the characteristic Péclet number of the reactor. Comparison between systems exhibiting low and high Péclet values has been offered, showing that systems in which diffusive mechanism is important, namely low Péclet number systems, are more adequate for the LSD technique.

In addition, the POD method has been revealed as an effective method to obtain the desired reduced model for the OPR, achieving a reduction up to the 50% of the computation time with respect to the FEM solved system. Besides, POD based dimensional reduction is efficient independently of the diffusive phenomenon relevance (i.e. for all the Péclet numbers values). It must be considered, too, that non-linearity present in the model certainly could appear in the solution in such way that the POD modes might significantly differ from the eigenfunctions of the Laplacian (Pinnau, 2008), and hence leading to different surrogate representations.

Finally, the obtained POD-based low representation of the system has been employed as the core of the optimal control strategy presented. Two different scenarios have been devised in order to achieve the desired objective: to start-up the OPR operation satisfying the safety constraints while maximising the reactants conversion. The first case consisted of a DO without restrictions, while the second one introduced constraints on the state variables. The utilisation of this ROM scheme has clearly decreased the computational times (around 1000 evaluations of the objective function are carried out in 30 seconds), while eased the system analysis.
Freeze-drying (lyophilisation) is a dehydration process well known in food industries as it is an operation which preserves the biological activity of thermosensitive components, as well as the organoleptic and nutritional properties of the material. It must be also mentioned that freeze-drying extends products shelf life, while making goods easier to transport and to store.

However, the slow drying rates and the working conditions (close to low absolute) constitute the main disadvantage of lyophilisation. This translates into an expensive process in terms of time and energy, and so economics. Thus, industrial uses of freeze-drying have been restricted to dehydration of high value products (Litchfield and Liapis, 1979; Pikal et al., 1983; Pikal, 2000; Trelea et al., 2007).

Three stages can be identified in the complete freeze-drying process (Song et al., 2002; Trelea et al., 2007), during which different physical phenomena take place, as depicted in Figure 5.1:

1. The first stage (freezing) involves a quick decrease of the sample temperature (reaching values below water triple point) in order to control the ice crystals size growth and to avoid possible damage to the material.

2. The second step, the so called primary drying, consists of heating of the sample under partial vacuum conditions (always below the triple point) to force ice sublimation. This leads to an interconnected porous structure which can be later rehydrated very effectively while preserving the organoleptic and nutritional properties of the product. During this stage (the longest one of the cycle), which
3. Finally, the last step, the secondary drying, is an ordinary drying process where the water still bounded to the porous matrix is desorbed by increasing the temperature. Typical figures for final moisture levels in the product are around 0.5% w/w.

The efficiency of the freeze-drying process in terms of productivity and product quality (obtaining of the highest quality in the shortest cycle time), is defined by various process variables (Trelea et al., 2007), being the product temperature the most determinant one. Although sublimation phenomenon is faster when temperature is increased, an overheating of the product could result into the collapse of the pore structure (Pikal and Shah, 1990) and the loss of the quality properties. This collapse phenomenon is usually associated with the glass transition temperature of the maximally freeze-concentrated phase. Thus, the proper handling of the product temperature will allow not only the maintenance of quality under market standard requirements but also to reduce process cycle time.

In this framework, theoretical modelling has been revealed as a very useful tool to achieve a better understanding of process dynamics and their influence on cycle time and product temperature history.

Classical freeze-drying references (Litchfield and Liapis, 1979; Millman et al., 1984; Pikal et al., 1983) offer an exhaustive description of the process dynamics, involving

Figure 5.1: Phase diagram for the freeze-drying phenomena.
5.2. System description

a high number of parameters. Still accurate, these models and their subsequent improvements e.g. (Boss et al., 2004; Bruttini et al., 1991; Mascarenhas et al., 1997), are computationally involved and time consuming, so unsuitable for on-line/real time control purposes. More recent publications echoed the need of developing simplified and control-oriented models. Trelea et al. (2007) proposed a lumped model for optimizing process performance. The inherent loss of dynamical information, which could affect the quality requirements of the final product, might be the main disadvantage of this model. The same motivation appears in Velardi and Barresi (2008), where two simplified models are developed for on-line applications. Although both models are based on the separation of slow and fast dynamics of the system in order to obtain the desired simplifications, no theoretical basis, and therefore systematics, is provided.

This chapter presents a first principle-based model, describing primary and secondary drying dynamics. By considering the corresponding thermophysical properties of the material, different characteristic times for freeze-drying dynamics are defined. Associated to these, the diverse time scales in which the involved physical phenomena take place are also revealed. From this rigorous time-scale analysis, a simplified distributed model is launched as a suitable tool not only to provide an accurate description of the freeze-drying dynamics but also to be used as the core for computing optimal operation policies.

5.2 System description

Porous media are defined as a permeated material constituted by an interconnected network of pores throughout which the vapour flux can circulate (Roth, 2012). Its importance is well known in many mass transfer applications, including freeze-drying of foods, where the process depends on the vapour water transport through a porous layer of dried solids (Bird et al., 2007). As a consequence, heat and mass transfer mechanisms will be considered in order to obtain a dynamic description of the freeze-drying process.

As commented in Section 5.1, freeze-drying dynamics has been extensively analyzed in literature (Bruttini et al., 1991; Litchfield and Liapis, 1979; Mascarenhas et al., 1997; Millman et al., 1985; Pikal et al., 2005; Ratti, 2001; Sadikoglu and Liapis, 1997). Relying on these references, a first principle based model describing the primary drying (stage which determines the product quality) and secondary drying is here presented. The main difference with respect to those other models is related to
Chapter 5

the treatment of the heat transfer phenomena in the dried layer. It must be remarked that separated energy transfer mechanisms for vapour and porous matrix in the dried region are considered.

For the sake of illustrative purposes, skim milk has been selected as the reference material. The physical parameters used to define the different work cases presented in this chapter, which have been taken from literature (Liapis and Bruttini, 1994; Mascarenhas et al., 1997; Millman et al., 1984), are detailed in Table 5.4.

![Freeze-dryer scheme](image)

**Figure 5.2**: Freeze-dryer scheme.

### 5.2.1 Assumptions

For modelling purposes the following standard assumptions are made (Mascarenhas et al., 1997; Millman et al., 1985; Trelea et al., 2007):

1. The frozen region has uniform heat and mass transfer properties.

2. There is a continuous interface (sublimation front) between the dried and the frozen region, with infinitesimal thickness.

3. At the interface, the concentration of water vapour is in equilibrium with ice.

4. The structure of the porous matrix is rigid provided that its temperature is below that of glass transition.
5.2. System description

5. The solid matrix is permeable, thus allowing the circulation of vapour fluxes.

5.2.2 Geometry

A generic system to be freeze-dried, as the one depicted in Figure 5.2 has been considered and approximated by the 1D slab presented in Figure 5.3. The porous dried media and a frozen region coexist separated by the sublimation front.

![Figure 5.3: 1D domain approximation for the freeze-drying system.](image)

Initially, the thickness of the dried region (and therefore the position of the front) is considered to be the 2% of the total length (Mascarenhas et al., 1997). Note that the existence of both regions will be needed for the boundary conditions to hold, avoiding discontinuities at \( x = 0 \) at the initial times of the operation.

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha_I )</td>
<td>Dried layer thermal diffusivity ([m^2/s])</td>
<td></td>
</tr>
<tr>
<td>( \alpha_{II} )</td>
<td>Frozen layer thermal diffusivity ([m^2/s])</td>
<td></td>
</tr>
<tr>
<td>( \alpha_v )</td>
<td>Vapour thermal diffusivity ([m^2/s])</td>
<td></td>
</tr>
<tr>
<td>( k_I )</td>
<td>Dried layer heat conductivity ([W/mK])</td>
<td></td>
</tr>
<tr>
<td>( k_{II} )</td>
<td>Frozen layer heat conductivity ([W/mK])</td>
<td></td>
</tr>
<tr>
<td>( \rho_I )</td>
<td>Dried layer density ([kg/m^3])</td>
<td></td>
</tr>
<tr>
<td>( \rho_{II} )</td>
<td>Frozen layer density ([kg/m^3])</td>
<td></td>
</tr>
<tr>
<td>( \rho_v )</td>
<td>Vapour density ([kg/m^3])</td>
<td></td>
</tr>
<tr>
<td>( K_m )</td>
<td>Dried material permeability ([m^2])</td>
<td></td>
</tr>
<tr>
<td>( \mu )</td>
<td>Vapour viscosity ([Pa\ s])</td>
<td></td>
</tr>
<tr>
<td>( v_{ref} )</td>
<td>Vapour reference velocity ([m/s])</td>
<td></td>
</tr>
<tr>
<td>( \sigma )</td>
<td>Stefan-Boltzmann constant ([W/m^2K^4])</td>
<td></td>
</tr>
<tr>
<td>( e_p )</td>
<td>Vapour chamber emissivity</td>
<td></td>
</tr>
<tr>
<td>( f_p )</td>
<td>View factor for the shelf radiation flux</td>
<td></td>
</tr>
<tr>
<td>( h_L )</td>
<td>Convective heat coefficient ([W/m^2K])</td>
<td></td>
</tr>
<tr>
<td>( \Delta H_s )</td>
<td>Sublimation enthalpy ([J/kg])</td>
<td></td>
</tr>
<tr>
<td>( L )</td>
<td>Sample length ([m])</td>
<td></td>
</tr>
<tr>
<td>( K_g )</td>
<td>Desorption coefficient ([1/s])</td>
<td></td>
</tr>
</tbody>
</table>
5.2.3 Mass transfer mechanisms

Mass vapour transport through the dried region is described by a modified continuity equation, which includes a desorption factor in terms of the product water content $C_b$. This is written as follows:

$$\varepsilon \frac{\partial \rho_v}{\partial t} = -\varepsilon \frac{\partial (\rho_v \vartheta)}{\partial x} - \rho_l \frac{dC_b}{dt}, \forall x \in (0, S(t)) \quad (5.1)$$

where $\varepsilon$ is the void volume fraction in the dried layer. Darcy’s law is used to compute the vapour flux velocity, reading $P_v$ for the vapour pressure:

$$\vartheta = -\frac{K_m}{\mu} \left( \frac{\partial P_v}{\partial x} \right) \quad (5.2)$$

Substituting (5.2) into (5.1) we get:

$$\varepsilon \frac{\partial \rho_v}{\partial t} = -\varepsilon \frac{\partial}{\partial x} \left( -\rho_v \frac{K_m}{\mu} \frac{\partial P_v}{\partial x} \right) + \rho_l \frac{dC_b}{dt}, \forall x \in (0, S(t)) \quad (5.3)$$

The above equation describes the motion of the vapour in the porous matrix.

5.2.4 Heat transfer mechanisms

When the heat transfer mechanisms present in the system are examined, three different transmission media must be considered: the porous layer, the vapour flux and the frozen material.

Regarding the porous layer, the heat balance accounts for heat conduction, bounded water sublimation heat and heat transfer between solid matrix and vapour:

$$\frac{\partial T_I}{\partial t} = \alpha_I \frac{\partial^2 T_I}{\partial x^2} + \frac{\Delta H_s}{C_{p_I}} \frac{dC_b}{dt} - \frac{h_{mv} S_{mv}}{\rho_l C_{p_I}} (T_I - T_v), \forall x \in (0, S(t)) \quad (5.4)$$

with $T_I$ being the temperature of the porous matrix and $h_{mv}$ the heat transfer coefficient. Noting the characteristic diameter by $d_p$, the contact surface between matrix and vapour can be defined as $S_{mv} = \frac{6(1-\varepsilon)}{d_p}$ (Mhimid et al., 2000).

Accordingly, for the vapour flux, heat transfer comprises conduction, advection, bounded water sublimation heat, and heat transfer between solid matrix and vapour, leading to:
5.3 Time-scale analysis

\[ \frac{\varepsilon \partial T_v}{\partial t} + \varepsilon \partial \frac{\partial}{\partial x} (T_v) = \varepsilon \alpha_v \frac{\partial^2 T_v}{\partial x^2} + \frac{\varepsilon h_{\text{mv}} S_{\text{mv}}}{C_p \rho_v} (T_I - T_v) - \rho I_D \Delta H_s C_p \rho_v \left( \hat{C}_b - C^\text{eq} \right) \]  
(5.5)

\[ - \rho I_D \Delta H_s C_p \rho_v + \rho I_D T_v \rho_v, \quad \forall x \in (0, S(t)) \]

with \( T_v \) being the vapour temperature.

Finally, throughout the frozen material, heat is assumed to be transferred only by conduction from the bottom shelf, so that:

\[ \frac{\partial T_{II}}{\partial t} = \alpha_{II} \frac{\partial^2 T_{II}}{\partial x^2}, \quad \forall x \in (S(t), L) \]  
(5.6)

5.3 Time-scale analysis

The inherent thermophysical properties of the system, this is, the thermal diffusivities, the desorption rate and the mass flux velocity, define a set of characteristic times in which the different physical phenomena take place. Associated to these, the different time scales involved in the freeze-drying process can be also described (see Table 5.1). Thus, aiming at achieving a better comprehension of the operation under study, the governing equations described in 5.2.3 and 5.2.4 are examined in terms of those different temporal scales. The utilisation of a dimensionless description of the system will facilitate such analysis. The new dimensionless field variables are listed in Table 5.2, and the following dimensionless space variable will be also employed:

\[ \xi = \frac{x}{L} \]  
(5.7)

This approach will allow us to focus just on the phenomenon of interest and to neglect mechanisms occurring at different time scales. For the freeze-drying case, the relevant time scale is the one related to the temperature distribution within the porous matrix. Working on this leading time scale, \( \Theta_I \), and after assuming local thermal equilibrium (LTE), process dynamics reduces to the following equations:

\[ \frac{d\hat{C}_b}{d\Theta_I} = -K_g L^2 \hat{\alpha}_I \left( \hat{C}_b - \hat{C}_b \right) \]  
(5.8)

\[ \frac{\partial \hat{\rho}_v}{\partial \Theta_I} = -L \hat{\rho}_v^{\text{ref}} \frac{\partial}{\partial \xi} \left( \hat{\rho}_v \hat{\rho}_v \right) - \frac{K_g L^2 \hat{\rho}_I C_b^{\text{ref}}}{\alpha_I \varepsilon \hat{\rho}_b^{\text{ref}}} \left( \hat{C}_b - \hat{C}_b \right) \]  
(5.9)
Chapter 5

Table 5.1: Characteristic times, together with their order of magnitude, and dimensionless time scales for the freeze-drying involved phenomena.

<table>
<thead>
<tr>
<th>Phenomena</th>
<th>Characteristic times</th>
<th>Order of magnitude</th>
<th>Dimensionless time scale</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass transfer:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vapor phase</td>
<td>$\tau_{vc} = \frac{L}{v_{ref}}$</td>
<td>milliseconds</td>
<td>$\Theta_{vc} = \frac{t_w}{L}$</td>
</tr>
<tr>
<td>Desorption</td>
<td>$\tau_b = \frac{1}{K_g}$</td>
<td>hours</td>
<td>$\Theta_b = tK_g$</td>
</tr>
<tr>
<td>Heat transfer:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Porous matrix</td>
<td>$\tau_I = \frac{L^2}{\alpha_I}$</td>
<td>minutes</td>
<td>$\Theta_I = \frac{t_{\alpha_I}}{L^2}$</td>
</tr>
<tr>
<td>Frozen region</td>
<td>$\tau_{II} = \frac{L^2}{\alpha_{II}}$</td>
<td>seconds</td>
<td>$\Theta_{II} = \frac{t_{\alpha_{II}}}{L^2}$</td>
</tr>
<tr>
<td>Vapour phase</td>
<td>$\tau_{v\alpha} = \frac{L^2}{\alpha_v}$</td>
<td>milliseconds</td>
<td>$\Theta_{v\alpha} = \frac{t_{\alpha_v}}{L^2}$</td>
</tr>
</tbody>
</table>

Table 5.2: Dimensionless field variables for the freeze-drying system.

\[
\frac{\partial \hat{T}_{II}}{\partial \Theta_I} = \frac{\alpha_{II}}{\alpha_I} \frac{\partial^2 \hat{T}_{II}}{\partial \xi^2}
\]  
(5.10)

\[
\frac{\partial \hat{T}_I}{\partial \Theta_I} = \frac{\partial^2 \hat{T}_I}{\partial \xi^2} - \frac{C_{p_v}^{ref} K_g L^2 \Delta H_s}{\alpha_I C_{p_v}(T_{ini} - T_L)} (C_{b_eq} - C_b)
\]  
(5.11)

where the dimensionless vapour flux velocity is defined as:

\[
\hat{\vartheta} = -\frac{P_v^{ref} K_m}{\vartheta^{ref} L \mu} \frac{\partial \hat{P}_v}{\partial \xi}
\]  
(5.12)
5.4. The matrix scale model

For the reference values given in Table 5.3, some of the terms in the RHS of Equations 5.8-5.11 can be neglected, what leads to the following simplified description:

\[
\frac{d\hat{C}_b}{d\Theta_M} \approx 0 \\
\frac{\partial(\hat{\rho}_v\hat{\vartheta})}{\partial \xi} \approx 0
\] (5.13)

\[
\frac{\partial \hat{T}_{II}}{\partial \Theta_I} \approx \frac{\alpha_{II}}{\alpha_I} \frac{\partial^2 \hat{T}_{II}}{\partial \xi^2} \\
\frac{\partial \hat{T}_I}{\partial \Theta_I} \approx \frac{\partial^2 \hat{T}_I}{\partial \xi^2}
\]

System 5.13 (together with the corresponding boundary conditions), when rewritten back in the original variables, is the model we referred to as the matrix scale model.

<table>
<thead>
<tr>
<th>Parameters of reference</th>
<th>Values</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>(C^v_{ref})</td>
<td>1674.7</td>
<td>J/kgK</td>
</tr>
<tr>
<td>(\rho^v_{ref})</td>
<td>1.54e^{-3}</td>
<td>kg/m^3</td>
</tr>
<tr>
<td>(k^v_{ref})</td>
<td>0.016</td>
<td>W/mK</td>
</tr>
<tr>
<td>(C^b_{ref})</td>
<td>0.22</td>
<td>kg water/kg solids</td>
</tr>
<tr>
<td>(\vartheta^v_{ref})</td>
<td>2.05</td>
<td>m/s</td>
</tr>
<tr>
<td>(L)</td>
<td>3</td>
<td>mm</td>
</tr>
</tbody>
</table>

Table 5.3: Reference values for the freeze-drying system.

5.4 The matrix scale model

By means of the performance of a rigorous time scale analysis, a simplification of the heat and mass coupled equations has been achieved. This approach has reduced the number of parameters involved, resulting into a simplified model which will be referred to as the matrix scale model. For the 1D case it is described as follows:
### Table 5.4: Input data and parameters for the freeze-drying time-scale model. PD = primary drying; SD = secondary drying. All temperatures are in kelvin; pressure is in Pascal units.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{pI}$</td>
<td>2595</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$\rho_I$</td>
<td>145</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$C_{pII}$</td>
<td>1967.8</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$\rho_{II}$</td>
<td>1058</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$k_I$</td>
<td>$8.826 \times 10^{-5} P_v + 2.706 \times 10^{-2}$</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$k_{II}$</td>
<td>2.4</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$K_g$</td>
<td>$11.08 \times 10^{-5}$</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$K_m$</td>
<td>$3.62 \times 10^{-10}$</td>
<td>(Liapis and Bruttini, 1994)</td>
</tr>
<tr>
<td>$\mu$</td>
<td>$8.36 \times 10^{-6}$</td>
<td>(Wang and Chen, 2005)</td>
</tr>
<tr>
<td>$f_p$</td>
<td>0.99</td>
<td>(Pikal, 2000)</td>
</tr>
<tr>
<td>$e_p$</td>
<td>0.94</td>
<td>(Pikal et al., 1983)</td>
</tr>
<tr>
<td>$\Delta H_s$</td>
<td>$2.79 \times 10^6$</td>
<td>(Mascarenhas et al., 1997)</td>
</tr>
<tr>
<td>$m_m(%)$</td>
<td>5.123</td>
<td>(Furmaniak et al., 2009)</td>
</tr>
<tr>
<td>$K_{GAB}$</td>
<td>0.9394</td>
<td>(Furmaniak et al., 2009)</td>
</tr>
<tr>
<td>$C_G$</td>
<td>5.512</td>
<td>(Furmaniak et al., 2009)</td>
</tr>
<tr>
<td>$T_g$</td>
<td>370</td>
<td>(Jouppila et al., 1997)</td>
</tr>
<tr>
<td>$T_{g2}$</td>
<td>138</td>
<td>(Jouppila et al., 1997)</td>
</tr>
<tr>
<td>$k_{GT}$</td>
<td>6.7</td>
<td>(Jouppila et al., 1997)</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>$5.6704 \times 10^{-8}$</td>
<td></td>
</tr>
<tr>
<td>$T_{ini}$</td>
<td>227</td>
<td></td>
</tr>
<tr>
<td>$T_c$</td>
<td>298</td>
<td></td>
</tr>
<tr>
<td>$T_L$</td>
<td>263 (PD)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>283 (SD)</td>
<td></td>
</tr>
<tr>
<td>$P_c$</td>
<td>10</td>
<td></td>
</tr>
</tbody>
</table>
5.4. The matrix scale model

5.4.1 Heat transfer equations

Heat transfer is governed by Fourier’s equation in both regions:

\[
\frac{\partial T_I}{\partial t}(x,t) = \alpha_I \frac{\partial^2 T_I}{\partial x^2}(x,t), \quad \forall x \in (0, S(t))
\] (5.14)

\[
\frac{\partial T_{II}}{\partial t}(x,t) = \alpha_{II} \frac{\partial^2 T_{II}}{\partial x^2}(x,t), \quad \forall x \in (S(t), L)
\] (5.15)

where \( T_I \) and \( T_{II} \) are the temperatures of the dried and the frozen layer, respectively, and \( S(t) \) denotes the position of the sublimation front. Over the product top \( (x = 0) \), radiation is the main heat transfer mechanism while at the bottom \( (x = L) \) convection must be considered (Bruttini et al., 1991). This leads to the following Neumann-type boundary conditions:

\[
k_I \frac{\partial T_I}{\partial x}(x,t) = \sigma e_p f_p (T_c^4 - T_I^4), \quad x = 0
\] (5.16)

\[
k_{II} \frac{\partial T_{II}}{\partial x}(x,t) = h_L (T_L - T_{II}), \quad x = L
\] (5.17)

where \( T_L \) is the shelf temperature and the convective heat transfer coefficient, \( h_L \), depends on the chamber pressure \( P_c \) (Bruttini et al., 1991; Sadikoglu and Liapis, 1997) as:

\[ h_L = 1.5358 P_c \]

Continuity of temperature across the front is also imposed:

\[ T_I = T_{II} = T_s, \quad x = S(t) \] (5.18)

It must be noted that the sublimation front constitutes a moving boundary, which calls for an extra boundary condition, the Stefan condition (Crank, 1987) at \( x = S(t) \), of the form:

\[
k_{II} \frac{\partial T_{II}}{\partial x}(x,t) - k_I \frac{\partial T_I}{\partial x}(x,t) = \Delta H_s (\rho_{II} - \rho_I) \frac{\partial S(t)}{\partial t}
\] (5.19)
5.4.2 Mass transfer equations

Due to the fact that heat and mass transfer mechanisms are strongly coupled, the continuity equation must be also defined in the dried region. For porous materials, the vapour velocity will be computed by Darcy’s law:

\[
\frac{\partial}{\partial x} \left( -\rho_v(x,t) \frac{K_m}{\mu} \frac{\partial P_v(x,t)}{\partial x} \right) = 0, \quad \forall x \in (0, S(t))
\]  
(5.20)

where the vapour density \( \rho_v \) is assumed to obey the Ideal Gas condition. In this way, vapour density can be calculated from \( P_v \) and \( T_i \) data directly. The other two parameters associated to Darcy’s equation are the matrix permeability, \( K_m \), and the vapour viscosity, \( \mu \).

Pressure value is fixed in the freeze-dryer chamber, while the sublimation phenomena origin a vapour flux from the front. This translates into the following Dirichlet and Neumann boundary conditions defined at \( x = 0 \) and \( x = S(t) \), respectively:

\[
P_v = P_c, \quad x = 0
\]  
(5.21)

\[
-\rho_v(x,t) \frac{K_m}{\mu} \frac{\partial P_v(x,t)}{\partial x} = \frac{\partial S(t)}{\partial t} (\rho_{II} - \rho_I), \quad x = S(t)
\]  
(5.22)

Since ice and vapour are in equilibrium at the moving front, once \( T_s \) is obtained by solving Equations (5.14)-(5.19), the Clausius-Clapeyron equation is employed to obtain \( P_v(S(t)) \), what closes the coupled mass transport problem. It should be remarked proportionality between gradient pressure and vapour flux in Equation (5.22), from which it is easy to see that, the driving force of the sublimation rate, and therefore drying, is the gradient pressure.

Despite the fact that the classical definition of freeze-drying distinguishes between primary and secondary drying as two consecutive stages, bounded water begins to be desorbed locally when the sublimation front passes by, creating a new portion of dried layer (Bruttini et al., 1991), what results into a coupled phenomenon. The evolution of water desorption in the product can be obtained from the vapour pressure in the dried region by solving the following Initial Value Problem (IVP), being the time dependence of the moisture content of the form (Mascarenhas et al., 1997; Millman et al., 1985; Pikal et al., 2005):

\[
\frac{dC_b}{dt}(t) = K_g \left( C_{b_{eq}} - C_b(t) \right), \quad \forall x \in [0, S(t))
\]  
(5.23)

\[
C_{b_{ini}} = C_b(x,0)
\]  
(5.24)
where $C_b$ represents the product water content (in kg water/kg solids) while $C_{b,eq}$ and $C_{b,ini}$ are the equilibrium and initial water contents of the sample, respectively. The equilibrium concentration for the sorbed water, $C_{b,eq}$, is given by the GAB equation (van den Berg and Bruin, 1981):

$$C_{b,eq} = \frac{m_m a_w C_G K_{GAB}}{(1 - a_w K_{GAB}) [1 + (C_g - 1) a_w K_{GAB}]}$$  \hspace{1cm} (5.25)

The water content of the mono-layer capacity $m_m$ is expressed in grams of water per 100 grams of solid. The water activity $a_w$ is here defined as the quotient between the vapour pressure in the dried layer, $P_v$, and the equilibrium vapour pressure at the considered temperature, given by the Clausius-Clapeyron equation, $P_{veq}$. The values employed for simulation purposes correspond to skim milk (Furmaniak et al., 2009).

### 5.5 Validation and numerical simulation of the matrix scale model

In order to verify the goodness of the simplified matrix scale model, a case study has been defined, consisting of the freeze-drying of a 3 mm and a 6 mm thickness. The governing equations for the matrix time scale were implemented in the commercial software COMSOL Multiphysics© and solved by the Finite Element method (FEM) together with an Arbitrary Lagrangian-Eulerian method (ALE) (Donea et al., 2004) in order to track the moving front (Mascarenhas et al., 1997).

The results obtained for the 3 mm thickness slab are presented in Figure 5.4, where trends for the product temperature distribution along the sample for the sublimation stage show good agreement with those reported in Mascarenhas et al. (1997) when the same boundary conditions are chosen. Differences might be attributed to the unlike initial conditions employed and to the use of inert gas in the literature case. In addition, final times for primary drying for both sample longitudes are compared in Table 5.5 to those reported in Mascarenhas et al. (1997) and Millman et al. (1985), showing also good agreement with literature.

Once stated the proper dynamic response of the developed matrix-scale model, it has been used to analyse the freeze-drying process of a 10 mm thick sample. For this new case, both sublimation and secondary drying stages have been simulated. The drying stage starts at the end of the sublimation phenomena. During this stage the whole sample can be considered dried. This new scenario is entirely governed by the
heat transfer mechanism corresponding to the dried material, so it can be described by Equation (5.14) and boundary conditions defined by Equations (5.16)-(5.17). The same numerical procedure employed in the validation case has been used to solve the 10 mm sample thickness case study, by using the data and parameters shown in Table 5.4.

<table>
<thead>
<tr>
<th>Sample length</th>
<th>Simulation</th>
<th>Mascarenhas et al. (1997)</th>
<th>Millman et al. (1985)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L = 3 , \text{mm}$</td>
<td>13.66 min</td>
<td>13.77 min</td>
<td>13.47 min</td>
</tr>
<tr>
<td>$L = 6 , \text{mm}$</td>
<td>54.83 min</td>
<td>55.26 min</td>
<td>54.07 min</td>
</tr>
</tbody>
</table>

Table 5.5: Primary drying times (in minutes): matrix scale model in comparison with literature results.

The product temperature profiles at different times can be seen in Figure 5.5. Both regions (dried and frozen) are well defined in the plot, being the lowest temperature point the one corresponding to the sublimation front. During the first 5-6 hours of the process a quick rise of temperature is detected, while a softer evolution follows next. Under conditions defined for this case (Table 5.4), the time needed to complete the primary drying was around 23.9 hours, being the secondary drying time of 6 hours.

Figure 5.6 shows the evolution in time for the temperature and position of the sublimation front during the primary drying stage. The behaviour of the front temperature follows the same trend as the whole product: an initial high increase, followed
5.5. Validation and numerical simulation of the matrix scale model

Figure 5.5: Product temperature profiles at different times for the skim-milk freeze-drying case during the sublimation stage.

Figure 5.6: a) Sublimation front position evolution and b) front temperature profile during primary drying for the skim-milk case.

by a smooth evolution. Regarding the front position, a quasi-linear evolution can be seen.

The water content profiles for different sample positions during the complete freeze-drying cycle are depicted in Figure 5.7. As reported in Furmaniak et al. (2009), Jouppila and Roos (1994) and Jouppila et al. (1997), Equation (5.25) is limited to a certain range of $a_w$ values. Numerically, this requires the adjustment of the water activities also in the simulations. From the results obtained, it is possible to determine the required freeze-drying cycle time required to fulfil a given quality specification for the product. In this case, an average moisture content ($C_{ave}$) below 0.02 kg water/kg solids at the end of the process has been selected, which corresponds
Figure 5.7: Profiles for the water content at different positions during the complete freeze-drying cycle. The horizontal line represents the final moisture requirement $C_b = 0.02 \text{kg water/kg solid}$.

to an average of 2% of water content. To attain such a product quality requirement, the process takes about 30 hours.

5.6 Conclusions

Conceptual modelling have been revealed as very useful tool to achieve a better understanding of freeze-drying process dynamics and their influence on cycle time and product temperature history, which are considered to be the key parameters for product quality, stability and productivity.

The proposed matrix scale model is a first principle based model describing coupled heat and mass transfer mechanisms, including also sublimation and desorption phenomena. It has been derived from a rigorous time-scale analysis, which has made use of the inherent thermophysical properties of the material, in order to define the different time scales involved in the operation.

The time-scale reduction approach has enabled to focus just on the phenomenon of interest, namely the time dependent product temperature distribution, and to neglect mechanism occurring at faster scales. Thus a simplified a low-dimensional control-oriented model has been obtained, being its performance validated by comparison with the existent literature.
Real time applications for the freeze-drying operational model

6.1 Introduction

The matrix scale model presented in Chapter 5 will be employed next as a suitable tool not only to provide an accurate description of the freeze-drying dynamics but also to be used as the core of further real time applications. In this on-line oriented framework, two different approaches will be presented.

Firstly, the computation of optimal operation policies for the freeze-drying process will be devised. The aim of the scheme is to easily and systematically determine optimal operation policies for freeze-drying processes, while trying to overcome disadvantages presented in previous literature. Drawbacks are mainly related to the use of computationally expensive dynamics models, parameter uncertainty (Litchfield and Liapis, 1982) or policies established on case-by-case basis (Alves and Roos, 2006; Sadikoglu et al., 1998).

In this chapter, the optimal control problem associated to the complete freeze-drying process will be simultaneously solved for primary and secondary drying stages. The time-scale based model has been defined as the core of the stated problem. It must be considered that if the mathematical model is not properly posed, this could lead once again to non-optimal process operation, which could result in a loss of product quality (Sadikoglu et al., 1998; Sadikoglu, 2005). Thus, the importance of a low dimensional control-oriented model.

Likewise, a new non-invasive methodology to estimate on-line the product temperature distribution in freeze-drying processes will be also presented. It makes use of a three-steps scheme which combines numerical simulation of the pressure tests with
Chapter 6

the performance of the *matrix scale* model and Reduced Order Methods (ROM). The estimates obtained by means of the simulated pressure rise tests (PRT) will feed the low-order representation of the freeze-drying drying dynamics. Numerical issues usually related to solving moving boundary problems, as it is the freeze-drying case, are overcome by applying the Landau transform (Landau, 1950; Crank, 1987; Illingworth and Golosnoy, 2005), which lead to a fixed reference frame. On the resulting work space, model reduction techniques, such as POD, will be used as a tool not only to simplify the system analysis but to minimize the computational effort as well.

6.2 Optimal control for freeze-drying processes

Aiming the establishment of optimal operations conditions, a number of methodologies have been describe in literature. The objective is to determine the operating conditions over the control variables, namely, the shelf temperature and the chamber pressure, which are the most intuitive controls. The purpose of these contributions is minimizing the drying time while satisfying the glass transition constraint (Gan et al., 2004; Rene et al., 1993). The suitable shelf temperature and chamber pressure conditions are frequently established on a trial-and-error basis (Alves and Roos, 2006). Thus, considering constant values for these variables arises as the simplest approach to determine optimal operation policies. Such approach normally results into non-optimal conditions for the primary drying (Liapis et al., 1996).

Since significant advantages can be obtained if shelf temperature and chamber pressure are varied during the process time (Pisano et al., 2010), the definition of a procedure to optimally determine time-varying operating profiles has become one of the scopes of recent research on freeze-drying (Velardi and Barresi, 2008; Pisano et al., 2010; Fissore et al., 2012; Pisano et al., 2013; Bosca et al., 2013a).

Finally, it must be mentioned that, in order to obtain a low dimensional non linear programming (NLP) problem, a Control Vector Parameterization (CVP) approach will be employed. The resulting NLP problem will be then efficiently solved by using a novel optimisation tool based on scatter-search (SSm).

6.2.1 Optimal control problem formalisation

Based on the *matrix scale model* previously introduced, diverse scenarios for the freeze-drying operation to be carried out can be devised, i.e. different pressure and/or
6.2. Optimal control for freeze-drying processes

Figure 6.1: Product temperature (solid line) and glass transition temperature (dotted line) at different positions during the complete freeze-drying cycle with constant \( T_L = 263 \, K \) (for primary drying), \( T_L = 283 \, K \) (for secondary drying) and \( P_c = 10 \, Pa \) profiles. The shelf temperature profile considered is represented on the upper left figure (dash-dotted line). The final moisture requirement \( C_{b,ave} = 0.02 \, kg \, water/\, kg \, solid \) is attained for a final cycle time of 29.888 h.

Such assessment has been formulated as a dynamic optimisation problem. The aim is to minimise the freeze-drying cycle time \( t_f \), while satisfying both product stability specifications \( C_{b,ave} \) as well as process dynamics. Stability conditions are related to the product temperature \( (T) \), which must be lower than the glass transition temperature \( (T_g) \) at any point of the sample during the whole freeze-drying cycle. Process dynamics is that described by equations (5.14)-(5.25) discussed before in Section 5.4, which is formally represented as \( f (\dot{x}, x, u, p, t) = 0 \). Mathematically, the problem can be stated as follows:

\[
\min_u t_f \tag{6.1}
\]

subject to:

\[
f (\dot{z}, z, u, p, t) = 0 \tag{6.2}
\]

\[
C_{b,ave} (t_f) = 0.02 \, kg \, water/\, kg \, solid \tag{6.3}
\]

\[
T (x, t) \leq T_g \tag{6.4}
\]
Figure 6.2: Product temperature (solid line) and glass transition temperature (dotted line) at different positions during the complete freeze-drying cycle with constant optimal control profiles for $T_L=251.75$ K and $P_c=25.398$ Pa. The final moisture requirement ($C_b^{\text{ave}}=0.02$ kg water/kg solids) is attained for a final cycle time of 38.45 h.

\[ 223 K \leq T_L \leq 323 K \quad (6.5) \]
\[ 10 \text{ Pa} \leq P_c \leq 60 \text{ Pa} \quad (6.6) \]

where $z \in \mathbb{R}^n$ are the states, $\dot{z}$ are their derivatives, $u \in \mathbb{R}^{nu}$ is the control vector that includes shelf temperature $T_L$ and chamber pressure $P_c$, and $p \in \mathbb{R}^{np}$ are a given set of parameters. The target average moisture content ($C_b^{\text{ave}}$) is considered as an endpoint constraint while the quality requirements ($T \leq T_g$) are taken into account as path constraints. As such, the latter will be introduced as penalties in the objective function. The upper and lower bounds for the considered controls have been chosen based on operational limitations of freeze-drying equipments.

It must be mentioned that the values of $T_g$ during the complete cycle were obtained by using the Gordon-Taylor equation (Gordon and Taylor, 1952):

\[ T_g = \frac{w_1 T_{g1} + k_{GT} w_2 T_{g2}}{w_1 + k_{GT} w_2} \quad (6.7) \]

Here, $T_g$ is the glass transition temperature of the sample, $w_1 = 1 - C_b$ and $w_2 = C_b$ are the weight fractions of matrix and water respectively. $T_{g1}$ and $T_{g2}$ are
6.2. Optimal control for freeze-drying processes

The glass transition temperatures of skim milk and water, and \( k_{GT} \) is a constant\(^1\). Parameter values corresponding to skim milk are taken from Jouppila et al. (1997).

For this system a constant shelf temperature and chamber pressure profile which leads to a cycle time \( (t_f) = 29.888 \) h and satisfies a final average moisture content not larger than 0.02 kg water/kg solids is presented in Figure 6.1. For this case, it is shown that at the beginning of the secondary drying, the product temperature is above that of glass transition. Such dynamic behaviour could lead to a collapsing phenomenon of the dried porous matrix and therefore, to rejection of the batch.

In order to overcome the drawbacks related to operate close to or over the collapse conditions, we propose the optimal control problem (6.1-6.6). As a first approximation, we consider the case of solving a simple NLP problem to determine optimal constant profiles along the whole operation time horizon for the decision variables \((T_L, P_c)\). Mathematically, this scenario is analogous to Problem (6.1-6.6), with \( u = [T_L, P_c] \) and \( T_{chamb} = 298 \) K. For this case, the value obtained for the optimal freeze-drying cycle time is \( t_f = 38.45 \) h. This value is a 28.6\% higher than the base case considered with \( T_L \) and \( P_c \) defined by the values referred to in Table 5.4. Such significant increase in operation time is the price to pay to avoid any violation of the temperature constraint (6.4) at any point of the sample and at any time. This

\(^1\)The temperature in this equation must be given in degrees Celsius
Figure 6.4: Product temperature (solid line) and glass transition temperature (dotted line) at different positions during the complete freeze-drying cycle with optimal control profile for $T_L$ and constant $P_c = 10$ Pa. The shelf temperature profile considered is represented on the upper left figure (dash-dotted line). The final moisture requirement ($C_{ave} = 0.02$ kg water/kg solids) is attained for a final cycle time of 31.64 h.

behaviour is shown in Figure 6.2, where the product and the glass transition temperature evolutions are represented at different sample positions. As a result, quality requirements for the final product are ensured but through penalizing the total process time. For this operational scenario, the associated water contents at different sample positions are depicted in Figure 6.3.

Next, variable control profiles will be considered to reduce the process time needed while preserving product stability and quality. A Control Vector Parametrisation approach (Vassiliadis, 1993; Vassiliadis et al., 1994) has been employed again to transform (6.1-6.6) into a non-linear optimisation problem (NLP) of low dimension, by employing the eSS-SSm Enhanced Scatter Search solver (Egea et al., 2009), following the same procedure than in Section 4.5.

One control variable case: shelf temperature

In this section, the same scenario analysed in previous works on freeze-drying optimisation (Trelea et al., 2007) is considered. The aim is to solve the NLP problem
6.2. Optimal control for freeze-drying processes

Figure 6.5: Profiles for the sample water content at different positions during the complete freeze-drying cycle with optimal control profile for $T_L$ and constant $P_c = 10$ Pa. The horizontal straight line represents the final moisture requirement ($C_{bave} = 0.02$ kg water/kg solids).

(6.1-6.6) when only one control variable (shelf temperature, $T_L$) is taken into account. Moreover, the operation time horizon is now unfixed, becoming the duration of the sublimation and drying stages two new decision variables. The associated optimisation problem is defined as follows:

$$\min_u t_f(\equiv t_{sub} + t_{dry})$$

subject to:

$$f(\dot{z}, z, u, p, t) = 0$$

$$C_{bave}(t_f) = 0.02 \text{ kg water/kg solid}$$

$$T(x, t) \leq T_g$$

$$223K \leq T_L \leq 323K$$

where now $u = [T_L, t_{sub}, t_{dry}] \in \mathbb{R}^{2\beta+2}$ and $P_c = 10$ Pa and $T_{chamb} = 298$ K are constant along the five intervals ($\beta = 5$) in which the sublimation and drying time horizons have been divided. In this case, the value obtained for the optimal cycle time is $t_f = 31.64$ h, which supposes a decrease of up to 17.71% when compared to the one attained for the optimal constant control variables case ($t_f = 38.45$ h). Note that no violations of the temperature constraint were detected (as shown in
Figure 6.6: Optimal $T_L$ profile for the scenario of one control variable.

Figure 6.4, fulfilling the desired quality properties of the final product. The water content evolutions at different sample positions and the optimal control profile for $T_L$ are presented in Figure 6.5 and Figure 6.6, respectively.

It must be noted that an increase of control discretization ($\beta$) would improve the freeze-drying operation time. However, such refinement could lead to a significant increase of the computational requirements to solve the problem. Consequently, a compromise between $\beta$ and computation costs must be achieved. After performing several tests, it was concluded that the selected level of discretization $\beta = 5$ is enough to clearly improve process time without the need of intensive computational efforts.

**Two control variables case: shelf temperature and chamber pressure**

Next, the influence of changes in chamber pressure over the freeze-drying cycle and the final time for the process will be analysed. To that purpose, two control variables, $T_L$ and $P_c$, are considered. The NLP problem (6.1-6.6) is now defined as:

$$\min_u t_f (\equiv t_{sub} + t_{dry})$$

subject to:

$$f (\dot{z}, z, u, p, t) = 0$$

$$C_{b_{\text{ave}}} (t_f) = 0.02 \text{ kg water/kg solid}$$

$$T (x, t) \leq T_g$$
6.2. Optimal control for freeze-drying processes

![Optimum temperature control profile](image)

Figure 6.7: Optimum temperature control profile obtained with SSm for the dynamic optimisation of a freeze-drying cycle for a skim-milk sample. Two control variables considered: $T_L$ and $P_c$.

\[ 223 \, K \leq T_L \leq 323 \, K \]  
\[ 10 \, Pa \leq P_c \leq 60 \, Pa \]

where $u = \{T_L, P_c, t_{sub}, t_{dry}\} \in \mathbb{R}^{43+2}$. Practical considerations regarding the operation of freeze-dryers on real food and biotechnological processing plants suggest the use of step changes (piecewise constant intervals for control discretization) over the chamber pressure ($P_c$). By comparing the water contents of the sample at different positions for the current case (Figure 6.8) with those ones obtained for the optimal constant controls ($T_L=251.75 \, K$ and $P_c=25.398 \, Pa$) scenario (Figure 6.5), a noticeable gain is obtained in terms of operation time. The cycle time for this optimal two control variables scenario is $t_f = 28.667 \, h$, which represents a significant reduction of $9.783 \, h (\approx 25.5 \%)$ and $2.973 \, h (\approx 9.40 \%)$ when compared with optimal constant profiles and one control variable ($T_L$) cases, respectively.

Shelf temperature profile (Figure 6.7) diminished due to self-cooling effect (Trelea et al., 2007) since ice sublimation rate decreases when the mass transfer resistance through the dry layer increases. During secondary drying, the moisture content of the product reduces, thus increasing the glass transition temperature in the product. As a consequence of those combined effects, the risk of collapse disappears since product temperature will be below $T_g$, leading to the possibility of increasing significantly the shelf temperature.

As expected, the profiles shown in Figure 6.7 and Figure 6.9 are quite smooth
Figure 6.8: Profiles for the sample water content at different positions during the complete freeze-drying cycle with optimal control profiles for $T_L$ and $P_c$. The horizontal straight line represents the final moisture requirement ($C_{wb}^{ave}=0.02$ kg water/kg solids).

during the primary drying, as compared with the secondary drying stage. This results into an easier implementation of those profiles in the equipments. Regarding temperature constraint violations no problems have been detected as shown in Figure 6.10. As a consequence, the porous structure and, therefore, quality of the final product is ensured while drastically reducing process time.

### 6.3 Non-invasive on-line estimation for freeze-drying processes

One of the most important factors to be considered in freeze-drying processes is product temperature (Trelea et al., 2007). An inconvenient over-heating of the sample could result into quality losses and consequent rejection of the batch. Thereby, its proper handling becomes critical, for it will lead not only to the desired product quality, but also to an enhancement on the operation efficiency by reducing the associated time and energy costs, as seen in Section 6.2.

Several attempts have been made to directly measure (Roy and Pikal, 1989; Hottot et al., 2005) or estimate (Kodama et al., 2013; Bosca and Fissore, 2011; Bosca et al., 2013b) this product temperature. However, those most relay on invasive methods, such as the insertion of thermocouples which, due to diverse disadvantages (i.e. the
6.3. Non-invasive on-line estimation for freeze-drying processes

Figure 6.9: Optimum pressure control profile obtained with SSm for the dynamic optimisation of a freeze-drying cycle for a skim-milk sample. Two control variables considered: \( T_L \) and \( P_c \).

risk of alterations in nucleation or crystal growth, damage to the product integrity and sterility issues (Velardi and Barresi, 2008), have been revealed unsuitable. This calls for new approaches based on non-invasive strategies.

It is in this new framework where techniques based on pressure rise methods, such as the Manometric Temperature Measurement (MTM) (Milton et al., 1997; Tang et al., 2005, 2006a,b,c), or the Pressure Rise Analysis (PRA) (Chouvenc et al., 2005; Velardi and Barresi, 2008; Barresi et al., 2009) have been arisen as a very attractive solution.

In this section, a new non-invasive methodology to estimate on-line the product temperature distribution in freeze-drying processes will be presented. It makes use of the combination of numerical simulation of the pressure tests with the performance of the matrix scale model and Reduced Order Methods (ROM).

6.3.1 The Rising Pressure Method

The importance of maintaining product temperature below the collapse values during primary drying is behind the motivation for developing non-invasive monitoring methods, among them such those based on the rising of the freeze-dryer chamber pressure.

In such tests, the condenser valve of the freeze-dryer is closed for a small period of time (usually 20-40 seconds). During this time interval, the vapour released by
Figure 6.10: Product temperature (solid line) and glass transition temperature (dotted line) at different positions during the complete freeze-drying cycle with optimal control profiles for $T_L$ and $P_c$. The shelf temperature profile considered is represented on the upper left figure (dash-dotted line). The final moisture requirement ($C_{ave}^{water}=0.02$ kg water/kg solids) is attained for a final cycle time of 28.667 h.

Sublimation passes throughout the dried region of the product and fills the freezing chamber, so increasing the pressure therein until equilibrium with the phase change front is reached (Tang et al., 2005; Velardi and Barresi, 2008).

Since devices are available in the chamber in order to measure pressure on-line, it is straightforward to obtain an estimate value for the front pressure and, subsequently, to recover the front temperature at the end of the test, $T_f$, by employing the Clausius-Clapeyron equation (Milton et al., 1997):

$$P_f = 133.3 \exp \left( \frac{-6144.96}{T_f} + 24.01849 \right)$$

(6.19)

**PRT simulations**

A model of the freeze-dryer chamber has been designed in order to allow numerical simulation of the pressure tests. This model consist of both material and energy balances, describing the evolution of the pressure, $P_c$ and chamber temperature, $T_c$, respectively, by means of the following expressions:
6.3. Non-invasive on-line estimation for freeze-drying processes

\[
\frac{dm_c}{dt} = \frac{A_s}{R_{FT}(S(t))} (P_f - P_c) \quad (6.20)
\]

\[
\frac{m_c T_c}{dt} = \frac{A_s}{R_{FT}(S(t))} (P_f - P_c) (T_f - T_c) \quad (6.21)
\]

where \(m_c\) notes the vapour mass, \(A_s\) the sublimation area and \(R_{FT}\) the mass resistance in the dried layer. This resistance must be experimentally determined for each material.

It must be remarked that Eqn 6.20 accounts only for inward flux since, during the PRT simulations, vapour is assumed to accumulate in the chamber. Figure 6.11 shows the evolution of chamber pressure, \(P_c\) compared with the sublimation front pressure, \(P_f\), and how those two values converge by the end of the essay, reaching equilibrium.

![Figure 6.11: Evolution of the chamber and front pressures during the simulation of one PRT.](image)

**6.3.2 Methodology description**

The proposed methodology comprises three steps, which are listed next:

- First, rising pressure experiments are carried out with the aim at providing front temperature estimations of quality.

- Next, the Landau transform is applied to the original moving boundary problem to obtain an equivalent system representation with fixed spatial domain. Here,
the estimates provided by the pressure experiments will be imposed as boundary conditions so to achieve the PDE system closure.

- Finally, the POD technique will be employed in order to reduce the dimensionality (and computational costs) of the transformed system without affecting the accuracy of the solution.

![Figure 6.12: Methodology scheme.](image)

**Step 1 - Front temperature estimation: PRT**

As explained above, estimations for the front temperature can be obtained by means of rising pressure experiments. Aiming at monitoring the whole primary drying, those tests must be performed at different times, (i.e. each hour). Nevertheless, since the front temperature evolves much faster during the first hour, a larger amount of experiments will be required during this early stage of the process in order to obtain proper estimates.

For the sake of validation, the same moving front problem was solved on the commercial FEM software (COMSOL Multyphysics©) by employing an adaptive remeshing method. The outcome values of $T_f$ are set side by side to the sublimation front temperature estimated by performing PRT in Figure 6.13.

Very good agreement for front temperature profiles is shown. Although, by the final part of the process (between 25 and 33 h) slight higher values are observed for the estimates. Two could be the causes behind these differences. One is the growing mass resistance (there is a longer dried region for the vapour to pass through) which creates a flux drop. Therefore, the time needed to reach equilibrium lengthens affecting the accuracy of the estimations. The other is related to the proximity of the heat source.
6.3. Non-invasive on-line estimation for freeze-drying processes

Figure 6.13: Evolution of the sublimation front temperature: real (solid line) vs. estimated values (asterisks).

placed at the bottom sample, which might influence the front temperature, too (Tang et al., 2005).

Step 2 - Handling the front: the Landau transform

In Stefan-like problems, such as freeze-drying process is, the front separating the frozen and dried zones is in motion, with variable velocity \( w \), due to the sublimation phenomenon taking place in between both regions.

As seen before in Chapter 3, several techniques can be applied to solve moving boundary problems like this one. From the point of view of the estimation scheme here proposed, the most interesting approach in order to deal with the moving front is the Landau transform (Landau, 1950; Illingworth and Golosnoy, 2005). This spatial transformation will allow to derive an equivalent mathematical description with fixed boundaries, and so, it will enable the application of reduced order methods, too.

For the sake of illustration, it is considered again the one-dimensional domain depicted in Figure 5.3, where the dried zone extends from the product top \( x = 0 \) to the front \( x = S(t) \), while the frozen region extends from the sublimation front \( x = S(t) \) to the product bottom \( x = L \).

By applying the coordinate transformation explained in 3.3.2, which for the case
Chapter 6

Here presented reads as:

\[ y = \frac{x}{S(t)} \]  \hspace{1cm} (6.22)

\[ z = \frac{x - S(t)}{L - S(t)} \]  \hspace{1cm} (6.23)

The moving boundary is fixed, since \( 0 \leq y, z \geq 1 \). In this coordinate system, new spatial and time operators are obtained, which applied to the heat problem defined by Eqns (5.14) and (5.15), results into:

\[
\frac{\partial T_I}{\partial \theta} = w \frac{y}{S(t)} \frac{\partial T_I}{\partial y} + \frac{\alpha_I}{S(t)^2} \frac{\partial^2 T_I}{\partial y^2}
\]  \hspace{1cm} (6.24)

\[
\frac{\partial T_{II}}{\partial \theta} = w \frac{1 - z}{(L - S(t))} \frac{\partial T_{II}}{\partial z} + \frac{\alpha_{II}}{(L - S(t))^2} \frac{\partial^2 T_{II}}{\partial z^2}
\]  \hspace{1cm} (6.25)

where \( \theta \) accounts now for the time in the transformed system.

The boundary conditions imposed by Eqns (5.16)-(5.19) will read as follows in the fixed coordinate system:

\[
\frac{k_I}{S(t)} \frac{\partial T_I}{\partial y}(0, \theta) = \sigma e_p f_p (T_4^I - T_4^I)
\]  \hspace{1cm} (6.26)

\[ T_I(1, \theta) = T_f \]  \hspace{1cm} (6.27)

\[ T_{II}(0, \theta) = T_f \]  \hspace{1cm} (6.28)

\[
\frac{k_{II}}{L - S(t)} \frac{\partial T_{II}}{\partial z}(0, \theta) - \frac{k_I}{S(t)} \frac{\partial T_I}{\partial y}(1, \theta) = \Delta H_s (\rho_{II} - \rho_I) \frac{\partial S(t)}{\partial \theta}
\]  \hspace{1cm} (6.29)

\[
\frac{k_{II}}{L - S(T)} \frac{\partial T_{II}}{\partial z} = \sigma e_b f_b (T_L^I - T_{II}^I) + h_L (T_L - T_{II})
\]  \hspace{1cm} (6.30)

The estimations of the front temperature provide by the PRT are employed in the boundary conditions above detailed as data, so the equations derived from the mass balance, and presented in Section 5.4.2, can be neglected. A complete summary of the notation employed is presented in Table 6.1.

**Step 3 - Model reduction: POD**

Model reduction is an important element of the current approach to the on-line estimation of product temperature distribution, for its combination together with PRT in an on-line methodology makes the difference with respect other PRT-based
6.3. Non-invasive on-line estimation for freeze-drying processes

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha_I$ Dried layer thermal diffusivity ($m^2/s$)</td>
<td>$2.36e^{-7}$</td>
</tr>
<tr>
<td>$\alpha_{II}$ Frozen layer thermal diffusivity ($m^2/s$)</td>
<td>$1.26e^{-6}$</td>
</tr>
<tr>
<td>$k_I$ Dried layer heat conductivity ($W/mK$)</td>
<td>$4.35e^{-2}$</td>
</tr>
<tr>
<td>$k_{II}$ Frozen layer heat conductivity ($W/mK$)</td>
<td>$2.4$</td>
</tr>
<tr>
<td>$\rho_I$ Dried layer density ($kg/m^3$)</td>
<td>$131.6$</td>
</tr>
<tr>
<td>$\rho_{II}$ Frozen layer density ($kg/m^3$)</td>
<td>$1048.6$</td>
</tr>
<tr>
<td>$K_m$ Dried layer permeability ($m^2$)</td>
<td>$4.43e^{-11}$</td>
</tr>
<tr>
<td>$\mu$ Vapour viscosity ($Pa s$)</td>
<td>$8.36e^{-6}$</td>
</tr>
<tr>
<td>$h_L$ Convective heat coefficient ($W/m^2K$)</td>
<td>$17.61$</td>
</tr>
<tr>
<td>$\Delta H_s$ Sublimation enthalpy ($J/kg$)</td>
<td>$2.79e^{0}$</td>
</tr>
<tr>
<td>$f_p, f_b$ View factor for the radiation fluxes</td>
<td>$0.99$</td>
</tr>
<tr>
<td>$\sigma$ Stefan-Boltzmann constant ($W/m^2K^4$)</td>
<td>$5.67e^{-8}$</td>
</tr>
<tr>
<td>$e_p$ Vapour chamber emissivity</td>
<td>$0.94$</td>
</tr>
<tr>
<td>$e_b$ Shelf emissivity</td>
<td>$0.28$</td>
</tr>
<tr>
<td>$L$ Sample length ($m$)</td>
<td>$0.01$</td>
</tr>
</tbody>
</table>

Table 6.1: Freeze-drying parameter values for the on-line estimation scheme.

methods presented in literature.

Considering the characteristics of the system under study, which exhibits non-homogeneous boundary conditions and a particular geometrical partition in two pieces, the most suitable ROM to be applied is the Proper Orthogonal Decomposition (POD).

Therefore, a set of temperature measurements must be obtained off-line in advance for both the frozen and dried regions, $T_I$ and $T_{II}$, respectively, by making use of the COMSOL simulation. These snapshots have been collected each 100 seconds, and the resulting data reservoir was employed to compute the empirical basis functions for each zone. Since the number of spatial nodes is much smaller than the number of snapshots, the direct method has been chosen to compute the corresponding POD basis by means of the the `matpod` function within the MatMOL toolbox.

Once the basis functions were available, the system equation described by Eqns (6.24)-(6.30) is projected onto the basis set, employing the energy captured by the system as reduction criterion (Sirovich, 1987). This leads to the low-dimensional ODE system to be solved, which in its most general form is described as follows:

$$\frac{dm_i}{dt} = Am_i + F, \quad i = I, II$$  \hspace{1cm} (6.31)
where $\mathcal{A}$ results from the projection of the diffusion and fictitious convection operators over the POD basis while $\mathcal{F}$ contains the non-homogeneous part of the boundary conditions.

![Graph](Image)

Figure 6.14: Comparison in terms of front position evolution between the COMSOL Multiphysics® (solid lines) and the values predicted by the proposed scheme (asterisks).

**Results**

First of all, and regarding the efficiency of the model reduction, it is important to mention that the COMSOL simulation uses 121 discretization points. Taking into account that two are the variables to be considered (temperatures in both regions), this makes a number of 242 ODEs to be solved together with the mesh displacement. On the other hand, 8 ODEs (4 for each zone) are employed by the POD-based reduced model, 30 times lower than the DOFs in the original system.

The evolution of the front position computed both using the COMSOL software and the estimation scheme proposed in this chapter is presented in Figure 6.14. For times below $25\text{h}$, the estimation values are in very good agreement with the COMSOL simulation results. Although, small differences ($\leq 5\%$) can be appreciated for times over that value, which is in accordance to the behaviour of the estimated front temperature presented in Section 6.3.2.

The same kind of conclusions can be drawn when examining Figure 6.15, where a comparison of the results in terms of temperature evolution at different points in the
6.4 Conclusions

In this chapter, the matrix scale model has been employed as the core of the proposed optimal control approach, which defines the operation conditions for minimizing freeze-drying cycle time while preserving product quality (final water content) through the solution of a dynamic NLP.

For the freeze-drying dynamics, diverse control scenarios have been analysed. First, constant optimal control profiles for $T_L$ and $P_c$ along the process time horizon were considered, overcoming temperature constraint violations that could lead to collapse of the product porous structure. For this scenario, final product quality is ensured at the expenses of an increase of about the 29% of the cycle time as compared with a standard non-optimal constant profile ($T_L = 263 \text{ K}$ and $P_c = 10 \text{ Pa}$).

After that, variable control profiles were proved to be successful in reducing process time while satisfying product quality standards. The resulting optimal profiles for $(T_L, P_c)$ led to reductions of up to an 17.71% (one control variable case - $T_L$) and

Figure 6.15: Comparison in terms of temperature evolution at different spatial points between the COMSOL Multiphysics® results (solid lines) and the values predicted by the proposed scheme (asterisks).

product is shown. A good correlation between both solutions is observed, but slight differences arise for the last points of the sample ($x = 8.3 \text{ mm}$), as a result again of the prediction errors at the end of the rising pressure experiments.
25.5% (two control variable case - $T_L$ and $P_c$) of the cycle time when compared with the case under optimal constant controls ($T_L = 251.75 \, K$ and $P_c = 25.398 \, Pa$).

In addition, a new non-invasive methodology for estimating on-line the product temperature distribution during the primary-drying of freeze-drying processes has been presented. This methodology also provides fair estimations for the front temperature and position.

On a three-steps basis, the proposed on-line scheme makes use of the so called Pressure Rise Tests (PRT) to obtain reliable estimates for the sublimation front temperature. In order to deal with the moving front, the Landau transform must be applied to the original moving boundary problem to obtain an equivalent system representation with fixed spatial domain. The estimates provided by the PRT step are employed here to obtain the closure of the PDE system formulated in the new working space. Finally, model reduction by means of the application of the Proper Orthogonal Decomposition (POD) technique is employed to decrease the dimensionality and lighten the computational costs of the transformed system.

In order to illustrate the suitability of the approach proposed, a simulation experiment was carried out. The results were compared to the accurate numerical solutions provided by a commercial FEM software (COMSOL Multphysics©) which employs an ALE method to solve the moving front problem. The estimates obtained are in good agreement with the COMSOL solutions. Slight differences can be appreciated at the final part of the process ($t > 30\, h$) because the time required to reach equilibrium increases in this period as a result of the vapour flux drop.

This procedure could be applied to higher dimensional spatial domains with complex geometries, where the benefits of employing reduced order models are expected to be further enhanced.
7.1 Introduction

In Chapter 3, the Enthalpy and Kirchhoff transformations were introduced as an advantageous procedure for solving Stefan-type moving boundary problems. This modelling approach can be encompassed into the so-called \textit{fixed-domain} family methods (Crank, 1987; Alexiades and Solomon, 1993). Such methods provide an alternative formulation of the problem, which applies over the whole fixed domain and where the Stefan condition is implicitly considered in the new governing equations (Crank, 1987).

Particularly, the Enthalpy-Kirchoff technique introduces two new extensive variables, $H(T)$ and $E(T)$ by means of the corresponding transformations from the temperature field, as defined by Eqn (3.30) and Eqn (3.31), respectively. The enthalpy variable $H(T)$ represents the total amount of energy of the system. It includes the existing jump between phases into its piecewise definition, what accounts for the Stefan condition in the reformulated system (see Appendix C for this justification). Besides, the discontinuity caused by the existence of different thermal conductivities can be successfully handle by means of the Kirchoff transform.

Although the combination of both transformations clearly eases the numerical solving of phase-change problems, this procedure has been scarcely employed in food industry Stefan problems. The available works in the related literature mostly deal with freezing or thawing (Voller and Cross, 1980; Mannapperuma and Singh, 1988; Fikiin, 1996; Comini and Saro, 1991; Alexiades and Solomon, 1993; Scheerlinck et al.,
2001; Santos and Lespinard, 2011), while there exists a lack for freeze-drying applications.

In this chapter, the Enthalpy-Kirchhoff approach will be employed to solve two different Stefan problems, both regarding phase change taking place at a constant temperature. Firstly, an illustrative example of a generic phase-change problem will be employed to exemplify the potential and advantages of the formulation in extensive variables, in both one-dimensional and two-dimensional domains. In addition, model reduction will be applied to the resulting systems, being the former one the core of the proposed reduced order dynamic observer.

The second problem presented in this chapter consists of the lyophilisation of a dairy product. Therefore, the freeze-drying matrix-scale model introduced in previous chapters will be here recovered, and solved for a multidimensional domain, too. The numerical procedures developed together with the results for one-dimensional and two-dimensional cases will be presented next.

Finally, it is worthy to remark that not only the application of the Enthalpy-Kirchhoff formulation to freeze-drying modelling, but also the application of model reduction to Stefan problems, and particularly in this extensive framework, are both thought to be original contributions of this thesis.

### 7.2 Generic phase change problem

As aforementioned, the first scenario to be presented in this chapter deals with a generic Stefan problem, which is characterised by an endothermic phase change taking place at constant temperature. The material considered can occur in two or more phases, and it is characterised by the data detailed in Table 7.1.

For a multidimensional domain \( \Omega \in \mathbb{R}^n \) with \( n = 1, 2 \), such problem could be mathematically defined as follows:

\[
\frac{\partial T_I}{\partial t}(\zeta, t) = \alpha_I \nabla T_I(\zeta, t), \quad \forall \zeta \in \Omega_I \quad (7.1)
\]

\[
\frac{\partial T_{II}}{\partial t}(\zeta, t) = \alpha_{II} \nabla T_{II}(\zeta, t), \quad \forall \zeta \in \Omega_{II} \quad (7.2)
\]

where \( \Omega = \Omega_I \cup \Omega_{II} \).

The Stefan condition, which determines the front velocity, can be expressed as
7.2. Generic phase change problem

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{p,l}$</td>
<td>2595</td>
<td>J/kgK</td>
</tr>
<tr>
<td>$\rho_l$</td>
<td>1000</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>$C_{p,ll}$</td>
<td>1967.8</td>
<td>J/kgK</td>
</tr>
<tr>
<td>$\rho_{ll}$</td>
<td>1058</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>$k_l$</td>
<td>2</td>
<td>W/mK</td>
</tr>
<tr>
<td>$k_{ll}$</td>
<td>2.4</td>
<td>W/mK</td>
</tr>
<tr>
<td>$\Delta H$</td>
<td>$2.79 \times 10^6$</td>
<td>J/kg</td>
</tr>
<tr>
<td>$T_{change}$</td>
<td>285</td>
<td>K</td>
</tr>
</tbody>
</table>

Table 7.1: Parameter values considered for numerical simulation of the generic phase change problem.

follows:

$$\Delta H (\rho_{ll} - \rho_l) \frac{\partial S}{\partial t} = k_{ll} \nabla T_{ll} \bigg|_{S_-} - k_l \nabla T_l \bigg|_{S_+}$$  \hspace{1cm} (7.3)

For a two-dimensional domain, by defining the front position as $y = S(x,t)$, as detailed in Appendix B, Eqn (7.3) turns into:

$$\frac{\partial S(x,t)}{\partial t} \Delta H (\rho_{ll} - \rho_l) = \left(k_{ll} \frac{\partial T_{ll}}{\partial y} - k_l \frac{\partial T_l}{\partial y}\right) \left[\left(\frac{\partial S(x,t)}{\partial x}\right)^2 + 1\right]$$  \hspace{1cm} (7.4)

where the front curvature is explicitly defined by the second factor of the RHS product.

The external boundary conditions have been imposed such that the sample is being heated by one (or more if 2D) boundaries, while the remaining edges are kept at temperature equal or lower than the phase change temperature $T_{change}$. Initial conditions have been accordingly chosen.

The application of Enthalpy and Kirchhoff transformations defined in Section 3.3.3 to the previous temperature-based equations leads to the general expression for the governing equation in the extensive formulation:

$$\frac{\partial H}{\partial t} = \Delta E, \quad \forall \zeta \in \Omega$$  \hspace{1cm} (7.5)

As seen in Chapter 3, the variable transformations defined by Eqn (3.30) and Eqn (3.31), corresponding to $H(T)$ and $E(T)$ respectively, result into following piecewise
definitions:

\[ H(T) = \begin{cases} 
H_{\text{ref}} + \rho_{\text{II}} C_{\text{pII}} (T_{\text{change}} - T_{\text{ref}}) + \rho_{\text{I}} C_{\text{pI}} (T - T_{\text{change}}) + \\
\quad + \Delta H (\rho_{\text{II}} - \rho_{\text{I}}) & \text{if } T > T_{\text{change}} \\
\in [H_{\text{ref}}, H_{\text{ref}} \Delta H (\rho_{\text{II}} - \rho_{\text{I}})] & \text{if } T = T_{\text{change}} \\
H_{\text{ref}} + \rho_{\text{II}} C_{\text{pII}} (T - T_{\text{ref}}) & \text{if } T < T_{\text{change}} 
\end{cases} \]

(7.6)

\[ E(T) = \begin{cases} 
E_{\text{ref}} + k_{\text{II}} (T_{\text{change}} - T_{\text{ref}}) + k_{\text{I}} (T - T_{\text{change}}) & \text{if } T > T_{\text{change}} \\
E_{\text{ref}} + k_{\text{II}} (T_{\text{change}} - T_{\text{ref}}) & \text{if } T = T_{\text{change}} \\
E_{\text{ref}} + k_{\text{II}} (T - T_{\text{ref}}) & \text{if } T < T_{\text{change}} 
\end{cases} \]

(7.7)

From these correspondences it is possible to obtain also the functions \( E(H) \) and \( H(E) \). Since the thermophysical properties are considered to be positive and bounded, and the temperature field is strictly monotonic, there exist a one-to-one mapping between intensive and extensive variables (Fikiin, 1996). Only dependences \( H(T) \) and \( H(E) \) exhibit a jump discontinuity at \( T = T_{\text{change}} \), which do not condition the numerical procedures since their inverse correspondences \( T(H) \) and \( E(H) \), which are the ones involve in the resolution algorithm, are indeed continuous. Accordingly, the initial, as well as the external boundary conditions, must be also transformed.

### 7.2.1 One-dimensional problem

For this one-dimensional case, a geometry consisting of a slab with length \( L = 0.01 \text{m} \) has been employed. The problem formed by Eqn (7.1) - Eqn (7.3) is completed by the following external boundary conditions:

\[ T_I(0, t) = T_c > T_{\text{change}} \]

(7.8)

\[ \frac{\partial T_{\text{II}}}{\partial x}(L, t) = 0 \]

After applying the above definitions to Eqn (7.8), as well as to the appropriate initial conditions, the corresponding extensive form of the those conditions read as follows:

\[ E(0, t) = E_c; \quad \frac{\partial E}{\partial x}(L, t) = 0 \]

(7.9)
7.2. Generic phase change problem

\[ H(x, 0) = H(T(x, 0)) \]  \hspace{1cm} (7.10)

Figure 7.1: Evolution at different process times for a) Enthalpy b) Kirchhoff variable when a N=41 nodes mesh is employed to solve a 1D generic Stefan problem.

The Finite Difference method has been employed to solve the system formed by Eqns (7.5), (7.9) and (7.10) considering a homogeneous spatial discretisation comprising \( N = 41 \) nodes. For the numerical solving, a 3-point-centered approximation for the spatial derivatives provided by the function `matfd` has been used. In addition, time integration of the resulting ODE systems has been carried out by means of the Matlab solver `ode15s`. Figure 7.1 shows the evolution of extensive variables along the sample, while in Figure 7.2 the evolution of the temperature field obtained by means of the Enthalpy-Kirchhoff method is compared to the results computed by using the Landau transform which, for the sake of validation, have been taken as the results of reference. Likewise, the advance of the moving front is compared in Figure 7.3.

7.2.2 Two-dimensional problem

In order to better describe the process in a two dimensional domain, the following assumptions are made:

1. The 2D domain is defined in a Cartesian system of coordinates.
2. There is a small region which has already changed of phase at the initial time.
3. The position of the moving front is given by \( y = S(x, t) \).
Figure 7.2: Evolution of the temperature along the product at different times when the Enthalpy-Kirchhoff method (solid) is employed to solve a 1D generic Stefan problem in comparison with the results of reference (marks).

The phase-change process is supposed to take place in a rectangular sample with length $L$ and width $2L$, so the spatial domain considered can be defined as $\Omega = \{(x, y) \in \mathbb{R}^2|0 \leq x \leq 2L, 0 \leq y \leq L\}$.

**Straight-shaped front**

As a first, and simple, approach, it has been assumed that the moving front is initially at a position $y_{ini} = S(x, 0)$, and that it preserves its shape all along the process time. The geometry employed for the two-dimensional case is the one depicted in Figure 7.4.

Considering the 2D domain, the external boundary conditions imposed, already in their extensive form, are defined as follows:

- **Side boundaries**
  \[
  \frac{\partial E}{\partial x}(0, y, t) = 0 \quad \text{and} \quad \frac{\partial E}{\partial x}(L_x, y, t) = 0
  \]  
  \[\text{(7.11)}\]

- **Top boundary**
  \[
  E(x, L_y, t) = E_c
  \]
  \[\text{(7.12)}\]

- **Bottom boundary**
  \[
  \frac{\partial E}{\partial x}(x, 0, t) = 0
  \]
  \[\text{(7.13)}\]

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7.2. Generic phase change problem

Once again, the system formed by Eqn(7.5) together with Eqns(7.11)-(7.13) and the suitable initial conditions has been solved by employing the Finite Difference method in a 21x41 nodes spatial grid with $\delta x = \delta y$.

A simple forward Euler method has been employed for time integration. For a 2D case like this is, the time step required to guarantee stability is defined by:

$$\delta t \leq \frac{1}{4}(\delta x)^2 \min \left( \frac{\rho_I C_{pI}}{k_I}, \frac{\rho_{II} C_{pII}}{k_{II}} \right)$$  \hspace{1cm} (7.14)

For the sake of validation purposes the 2D results obtained have been compared with those corresponding to the 1D case presented in Section 7.2.1, as can be seen in Figure 7.5 a), since due to the boundary conditions chosen for the multidimensional case, both systems can be considered analogous. A very good agreement between both methods is observed. Complementary, the advance of the moving front is shown in Figure 7.5 b).

Irregular front

In this example, a system configuration which results into a front exhibiting an irregular shape (curvature) has been considered. To that purpose, the boundary conditions imposed on the external boundaries have been slightly modified, adding a new isolated edge, as described next:
Figure 7.4: Two-dimensional domains to be considered in the numerical solving of the generic Stefan problem, together with the corresponding initial and boundary conditions imposed.

Figure 7.5: Two-dimensional validation results: a) Enthalpy-Kirchhoff method in 2D (marks) in comparison with analogous 1D method results (solid) b) Front evolution at different process times for the 2D generic with straight front.

- Side boundaries
  \[ E(0, y, t) = E_c \quad \frac{\partial E}{\partial x}(L_x, y, t) = 0 \]  
  \[(7.15)\]

- Top boundary
  \[ E(x, L_y, t) = E_c \]  
  \[(7.16)\]

- Bottom boundary
  \[ \frac{\partial E}{\partial x}(x, 0, t) = 0 \]  
  \[(7.17)\]
7.3 Freeze-drying problem

The second application of the Enthalpy-Kirchhoff method deals with the so-called primary drying of freeze-drying processes. Therefore, the operational model presented...
in Chapter 5 is here recovered again and employed as the core of the proposed problem.

Since sublimation is assumed to happen at constant temperature, no pressure equations will be considered, and therefore, only the thermal problem will be taking into account. This results into a model with the suitable structure for the Enthalpy-Kirchhoff transformations to be exploited.

The freeze-drying system configuration involves external boundary conditions such that the temperature at the product edges is always higher, or at least equal, than the sublimation temperature $T_s = 260 \, K$. Consequently, the moving front temperature will make up for the minimum of the system.

Therefore, this scenario will be characterised by the non-monotonic nature of variables $T$, and hence $H(T)$ and $E(T)$, too. This will constitute a key issue when the mappings from intensive ($T$) to extensive variables ($E$ and $H$) will be defined.

The corresponding governing equation, formulated in extensive variables, will take the form of Eqn (7.5). The mathematical description of problem to be solved will be completed once the initial and external boundary conditions were defined. Following the structure of the previous section, these conditions will be presented together with the formalisation of both one-dimensional and two-dimensional cases.

Finally, it must be mentioned that, again in this chapter, the numerical values employed for simulation purposes are those referred to in Table 5.4 for a skim-milk product.

### 7.3.1 One-dimensional case

A slab with length $L = 0.01 \, m$ is freeze-dried by imposing constant temperatures $T_c = 303 \, K$ and $T_L = 266 \, K$ on boundaries $x = 0$ and $x = L$, respectively. Complementary, the phase change temperature has been set to $T_s = 260 \, K$. Note since $T_c, T_L > T_s$ the sublimation front temperature $T_s$ constitutes the minimum value of the system.

This system configuration prevents from defining a one-to-one mapping between temperature and the extensive variables. As can be seen in Figure 7.7, each value of $H$ and $E$ corresponds to a unique value of $T$. However, the opposite does not hold: two different values of $H$ and $E$ could lead to the same temperature value.

To tackle numerical solving, a slight modification of the transformations definition must be firstly adopted. This variation refers to the change criterion defined for telling dried from frozen region. Instead the usual phase-change temperature $T_s$ employed in Eqn (7.6) and Eqn (7.7), it will be taken into account an approximated front position.
7.3. Freeze-drying problem

\[ H(T) = \begin{cases} 
H_{ref} + \rho_{II} C_{p_{II}} (T_s - T_{ref}) + 
\rho_{II} C_{p_{I}} (T - T_s) + \Delta H_s (\rho_{II} - \rho_I) & \text{if } x > \tilde{S}(t) \\
\in [H_{ref}, H_{ref} \Delta H_s (\rho_{II} - \rho_I)] & \text{if } x = \tilde{S}(t) \\
H_{ref} + \rho_{II} C_{p_{II}} (T - T_{ref}) & \text{if } x < \tilde{S}(t) 
\end{cases} \] (7.18)

\[ E(T) = \begin{cases} 
E_{ref} + k_{II} (T_s - T_{ref}) + k_{I} (T - T_s) & \text{if } x > \tilde{S}(t) \\
E_{ref} & \text{if } x = \tilde{S}(t) \\
E_{ref} + k_{II} (T - T_{ref}) & \text{if } x < \tilde{S}(t) 
\end{cases} \] (7.19)

Now, it is possible again to compute \(T(H)\) and \(T(E)\), as well as \(H(E)\) and \(E(H)\), functions that will be employed further in the numerical solving of the problem. Besides, by means of the above definitions, the boundary conditions expressed in terms

\[ \tilde{S}(t). \] According to this, functions \(H(T)\) and \(E(T)\) for the current freeze-drying scenario are defined as follows:

Figure 7.7: Representative extensives variables for scenario 2 a) \(H(T)\) b) \(E(T)\). Note the boxes delimit the temperature ranges which can belong to both phases, preventing the one-to-one mapping between temperature and the extensive variables.
of the extensive variables read:

\[ E(0, t) = E(T_c) = E_c; \quad E(L, t) = E(T_L) = E_L \]  

(7.20)

Thus, given the suitable initial conditions, numerical calculations have been carried out by following the next outlined algorithm:

1. Obtain \( H_{ini} \) and \( E_{ini} \) from \( T_{ini} \) by means of Eqn (7.18) and Eqn (7.19), respectively.

2. Solve system formed by Eqns (7.5) and (7.20).

3. Identify the node corresponding to the lowest value of \( H(E) \) in the dried region (upper branch in the \( H \) vs. \( T \) graph).

4. Define \( \tilde{S}(t) \) as the subsequent spatial node.

5. Compute \( E(H) \) by Eqn (7.19)

6. Repeat steps 2-6 until the final process time is reached.

For the sake of algorithm testing, implicit and explicit Finite Difference schemes, consisting of a 3 point-centered space stencil, have been employed in step 2, both achieving high accuracy. However, it must be mentioned that the computational times required by the implicit scheme, which makes use of the matlab solver \texttt{ode}15\texttt{s}, were larger than the times corresponding to the explicit implementation, where a forward Euler method was used.

The results obtained for the implicit scheme, in comparison with those corresponding to the temperature-formulated problem are shown in Figure 7.8. Likewise, the explicit implementation outcomes are depicted in figure 7.9.

In addition, the evolution in time of the real front position has been obtained a posteriori by numerical interpolation of the known value \( T_s \). For the sake of comparison and results validation, the front evolution in time obtained has been plotted together with the results corresponding to solve the same problem formulated in intensive variables by employing the Landau method in Figure 7.10. Note the good agreement between both, as well as the characteristic stepwise form of the Enthalpy-Kirchhoff result.
7.3. Freeze-drying problem

Figure 7.8: Implicit numerical scheme results: a) Comparison between the results obtained through the Enthalpy-Kirchhoff formulation (solid) at different freeze-drying process times and the temperature-formulated results (marks). b) Different time profiles for the Enthalpy variable $H(T)$.

Figure 7.9: Explicit numerical scheme results: a) Comparison between the results obtained through the Enthalpy-Kirchhoff formulation (solid) at different freeze-drying process times and the temperature-formulated results (marks). b) Different time profiles for the Enthalpy variable $H(T)$.

7.3.2 Two-dimensional case

The algorithm presented before will be employed next to solve the multidimensional extension of the freeze-drying problem presented in Section 7.3.1. For this example it has been considered again a moving front which preserves its shape along the process. Besides, the assumptions exposed in Section 7.2.2 regarding 2D cases hold for the current example study, too. For the sake of illustration, the spatial domain considered has been depicted in Figure 7.11.

Therefore, the two-dimensional freeze-drying problem to be solved is described
Figure 7.10: Comparison between the front position obtained through the Enthalpy-Kirchhoff based numerical algorithm (solid) and the front position computed by a temperature-base front-fixing method (dashed): a) implicit scheme b) explicit scheme

by the governing equation defined in Eqn (7.5) together with the following boundary conditions:

- Side boundaries
  \[ \frac{\partial E}{\partial x}(0, y, t) = 0; \quad \frac{\partial E}{\partial x}(L_x, y, t) = 0 \]  
  (7.21)

- Top boundary
  \[ E(x, L_y, t) = E_c \]  
  (7.22)

- Bottom boundary
  \[ E(x, 0, t) = E_L \]  
  (7.23)

The system is closed by the corresponding initial conditions, expressed in terms of the enthalpy variable:

\[ H(x, y, 0) = H(T(x, y, 0)) = H_{ini}(x, y) \]  
(7.24)

In this case, a grid of 21x41 nodes has been considered, which is equally spaced in both directions. Likewise, a FTCS scheme has been chosen in order to carry out the numerical resolution of the problem. As in previous examples, such scheme consists of a 3-point-centered stencil to approximate the spatial derivatives, together with a forward Euler method for time integration.

The results obtained have been compared, for the sake of validation, with those
7.3. Freeze-drying problem

Figure 7.11: Two dimensional domain, together with the imposed boundary conditions, for the 2D case of the freeze-drying scenario 2.

ones corresponding to the 1D case presented in Section 7.3.1, since both cases can be considered analogous. As can be seen in Figure 7.12, an excellent agreement with the reference system have been achieved.

Moreover, the distribution of the temperature along the sample at different operation times is presented in Figure 7.13. Likewise, the evolution of the moving front in time is shown in Figure 7.14.
7.4 Model reduction

In this section, the obtaining of low dimensional representations of the proposed models for solving Stefan problems will be tackled. In the choice of the technique employed to obtain the corresponding reduced systems, the structure of the freeze-drying problem formulated in extensive variables (Eqn (7.5)) plays a key role. Since it consists of the Laplacian operator without non-linear terms, the Laplacian Spectral Decomposition (LSD) emerges as the most suitable reduction method.

However, the application of the LSD technique calls for homogeneous boundary conditions to be imposed. Therefore, a transformation must be applied to the problem equations in order to meet this requirement (Courant and Hilbert, 1953; Balsa-Canto et al., 2004a):

\[
\bar{H}(\zeta, t) = H(\zeta, t) - H_c \tag{7.25}
\]

\[
\bar{E}(\zeta, t) = E(\zeta, t) - E_c \tag{7.26}
\]

Note that both extensive variables have been transformed. Therefore, the new governing equation reads as follows:

\[
\frac{\partial \bar{H}}{\partial t}(\zeta, t) = \Delta \bar{E}(\zeta, t) \tag{7.27}
\]
Figure 7.13: Temperature distribution for the two-dimensional problem corresponding to freeze-drying scenario2: a) Initial time b) 1/3 final time c) 2/3 final time d) Final process time.

The new system will be completed with corresponding boundary conditions, which in a general form, they can be expressed as:

- Dirichlet type conditions
  \[ \bar{E}(\Gamma, t) = 0 \]  
  \[ (7.28) \]

- Neumann conditions
  \[ \nabla \bar{E}(\Gamma, t) = 0 \]  
  \[ (7.29) \]

Together with the suitable initial conditions. Likewise, the correspondences between intensive and extensive variables will reflect this transformations, too, leading to new functions \( \bar{H}(T), \bar{E}(T), T(\bar{H}), T(\bar{E}), \bar{H}(E) \) and \( \bar{E}(\bar{H}) \).
Figure 7.14: Evolution in time of the sublimation front for the 2D case of the freeze-drying problem.

### 7.4.1 The Gibbs phenomenon

As previously commented in Chapter 2, the Laplacian Spectral Decomposition (LSD) approximates the exact solution of a given PDE by considering a discrete version of the original problem, and expressing its solution as a truncated Fourier series (Beckermann et al., 2007) of the form of Eqn (2.3). Usually, they yield to highly accurate approximations. However, if there exist a jump discontinuity in the state variables, then spurious oscillations appear as the discontinuities are approached, dropping convergence as well as accuracy of the method. These oscillations are the manifestation of the so called Gibbs phenomenon (Gottlieb and Orszag, 1977).

According to this, and since the variable transformation defined in Eqn (7.25) preserves the jump discontinuity of the enthalpy variable, those characteristic oscillations will also show up when a LSD-based model reduction scheme is applied to the Enthalpy-Kichhoff formulation.

For the sake of illustration, an off-line implementation of the LSD method has been carried out by employing the enthalpy results corresponding to the problem solved in Section 7.2.1, which have been firstly transformed according to Eqn (7.25). The comparison between FD and LSD-based outputs is shown in Figure, where the oscillations due to the Gibbs phenomenon can be clearly seen.
7.4. Model reduction

Figure 7.15: Comparison between the Finite Differences results (solid) and the LSD results for two different approximations: with 31 terms (dashed) and 11 terms (dot) obtained for $H(T)$. Note that the less the terms in the truncated serie, the higher are the effects of the Gibbs phenomenon.

**Smoothing approach**

In order to reduce the effects of the Gibbs phenomenon, the straightforward alternative consists of considering that the phase change can occur among a small range of temperatures $\Delta T_{\text{change}} = [T_{\text{change}} - \delta T, T_{\text{change}} + \delta T]$ (Scheerlinck et al., 2001). Once this assumption is introduced into Eqn (7.6) the following continuous function is obtained:

$$H(T) = \begin{cases} 
H_{\text{ref}} + \rho_{II}C_{pII} (T_{\text{change}} - T_{\text{ref}}) + \\
+ \rho_I C_{pI} (T - T_{\text{change}}) + \Delta H (\rho_{II} - \rho_I) & \text{if } T > T_{\text{change}} + \delta T \\
H_{\text{ref}} + m(T - T_{\text{ref}}) + a & \text{if } T \in \Delta T_{\text{change}} \\
H_{\text{ref}} + \rho_{II}C_{pII} (T - T_{\text{ref}}) & \text{if } T < T_{\text{change}} - \delta T 
\end{cases}$$  

(7.30)

where $m$ and $a$ define the slope and the y-intercept, respectively, of the straight line now defining $H(T)$ at the phase change range.

Accordingly, before defining the corresponding smooth Kirchhoff function, it is needed to introduce a piecewise thermal conductivity of the form:
Figure 7.16: Comparison between the discontinuous Enthalpy function and the corresponding smooth one obtained with $\delta T = 3.5 K$.

\[
k(T) = \begin{cases} 
    k_I & \text{if } T > T_{\text{change}} + \delta T \\
    m_k(T - T_{\text{ref}}) + c & \text{if } T \in \Delta T_{\text{change}} \\
    k_{II} & \text{if } T < T_{\text{change}} - \delta T 
\end{cases} 
\]

(7.31)

Now, since the thermal conductivity is a function of temperature, it should be differentiated respect to $T$ to obtain the smooth piecewise definition of $E(T)$:

\[
E(T) = \begin{cases} 
    E_{\text{ref}} + 0.5m_k \left[\left(T_{\text{change}} + \delta T\right)^2 - T_{\text{ref}}^2\right] + \\
    + \left(c - m_k T_{\text{ref}}\right) \left((T_{\text{change}} + \delta T) - T_{\text{ref}}\right) + \\
    + k_I(T - T_{\text{change}}) & \text{if } T > T_{\text{change}} + \delta T \\
    E_{\text{ref}} + 0.5m_k(T^2 - T_{\text{ref}}^2) + \left(c - m_k T_{\text{ref}}\right)(T - T_{\text{ref}}) & \text{if } T \in \Delta T \\
    E_{\text{ref}} + 0.5m_k \left[\left(T_{\text{change}} - \delta T\right)^2 - T_{\text{ref}}^2\right] + \\
    + \left(c - m_k T_{\text{ref}}\right) \left((T_{\text{change}} - \delta T) - T_{\text{ref}}\right) + \\
    + k_{II}(T - (T_{\text{change}} - \delta T)) & \text{if } T < T_{\text{change}} - \delta T 
\end{cases} 
\]

(7.32)

A noticeable improvement in the accuracy of the reduced system has been achieved
7.4. Model reduction

when the smooth functions previously defined were considered, specially for the case which employs 31 modes, as can be seen in Figure 7.17.

![Graph showing comparison between Finite Differences and LSD results](image)

Figure 7.17: Comparison between the Finite Differences results (solid) and the LSD results for two different approximations: with 31 terms (dashed) and 11 terms (dot) obtained for the smooth \( H(T) \) with \( \delta T = 3.5K \). Note that the less the terms in the truncated serie (i.e. neig), the higher are the effects of the Gibbs phenomenon.

However, and despite of the smoothing approach, the Gibbs phenomenon has not been completely removed from the system, as shown the spurious oscillations are still important for the case employing 11 modes.

### 7.4.2 Results

The LSD method has been combined with the smoothing approach in order to obtain the low-dimensional representation of the system presented in Section 7.2. Both, one-dimensional and two-dimensional cases, have been considered, and they will be presented next.

#### One-dimensional case

The LSD technique has been applied in order to solve the system described by Eqns (7.27)-(7.29). This is the equivalent system, with homogeneous boundary conditions, to the one before presented in Section (7.2.1).

The numerical solving has been carried out by considering 17 modes in the reconstruction of the enthalpy variable, while for the time integration of the resulting ODE system a forward Euler method has been employed.
Figure 7.18: Comparison between the results obtained when employing the LSD method with \( neig = 17 \) and \( \delta T = 3.5K \) (dashed) and the results for the full model (solid), also considering the smoothing approach for: a) reconstructed function \( \tilde{H}(T) \) b) the corresponding temperature field \( T(\tilde{H}) \).

In Figure 7.18 a) the reconstructed enthalpy variable with smoothing \( \tilde{H} \) is compared to the results obtained for the full smooth model (i.e. FD results). It can be clearly noted that the higher oscillations in the results are related to the initial times. This inaccuracy will be subsequently transmitted to the temperature field \( T \), as can be seen in Figure 7.18 b), since the function \( T(\tilde{H}) \) is employed to recover the system results in intensive variables.

Aiming at refining this LSD approximation as well as minimising the associated error to the oscillations, a filter relaying on the mean values of the reconstructed field \( \tilde{H} \) around the jump discontinuity has been designed. The results obtained, once the intensive variable has been recovered, are shown in Table 7.2. In this table, the model reduction results employing the smoothing approach, both with and without filter, are compared in terms of the relative error committed with respect to the full model with smoothing.

<table>
<thead>
<tr>
<th>Method</th>
<th>Max. rel. error(%)</th>
<th>Mean rel. error(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smoothing</td>
<td>1.65</td>
<td>0.21</td>
</tr>
<tr>
<td>Smoothing with filter</td>
<td>1.42</td>
<td>0.19</td>
</tr>
</tbody>
</table>

Table 7.2: Results of the LSD method: relative errors for the 1D case when 17 modes were employed for the reconstruction. Data are referred to the intensive variable \( T(\tilde{H}) \)

Finally, it must be mentioned that, these new smooth systems (full or reduced) diverge from the original one as the phase change range \( \Delta T \) is widen. Thus, a compromise between reduction and accuracy must be found.
7.4. Model reduction

Two-dimensional case

Two-dimensional case has been solved by following the same strategy: firstly, the static procedure was implemented, and once it was available, the dynamic simulations for the reduced model were then carried out.

Reduced representations have been obtained for systems exhibiting either straight or irregular phase-change fronts. Next, results will be shown in Table 7.3.

<table>
<thead>
<tr>
<th>Case</th>
<th>Number of modes</th>
<th>Max. rel. error(%)</th>
<th>Mean rel. error(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plane front no filter</td>
<td>115</td>
<td>2.06</td>
<td>0.20</td>
</tr>
<tr>
<td>Plane front with filter</td>
<td>115</td>
<td>1.93</td>
<td>0.18</td>
</tr>
<tr>
<td>Curve front</td>
<td>301</td>
<td>3.00</td>
<td>0.18</td>
</tr>
</tbody>
</table>

Table 7.3: Results of the LSD method: 2D case, relative errors for the intensive variable $T(\tilde{H})$ when compared with the full model.

The reduction obtained when the regular front is considered is higher than the 80%, since the full model mesh consisted of 861 nodes and now only 115 modes are being used in the reconstruction. When the irregular front system was simulated, in order to achieve the same magnitude of mean error than in the other 2D cases, more modes were needed to be considered. Nevertheless, the reduction is important yet, of around 65%.

Figure 7.19: LSD results (marks) in 2D for a plane front system at when compared with the full model (solid): a) reconstructed enthalpy evolution in time. b) reconstructed sample temperature evolution in time.

7.4.3 Dynamic observer

As already pointed out, and due to the discontinuity exhibited by the enthalpy variable, the application of the LSD method to Stefan-type problems depends on the
approximation of the energy jump caused by the phase change so to obtain a continuous enthalpy variable. In this way, the adverse effects of the Gibbs phenomenon can be diminished.

However, this approach has a main drawback, which is directly related to the value assigned to the new phase change interval $\Delta T$. The wider this temperature range, the greater the differences between smooth and no smooth systems, and thus the error. This situation calls for a new methodology capable of providing an accurate field reconstruction while taking advantage of the developed ROM scheme.

Assuming that the nature of the system under study is suitable for the obtaining of a given number of measurements, an alternative approach to field reconstruction is proposed. Such methodology involves the design of a reduced-order dynamic observer. Following the strategy proposed by Alonso et al. (2004), the reconstruction of the distributed extensive field will be carried out by employing those measurements to estimate the unobserved states in combination with the projection of the observer onto the LSD basis functions.

Figure 7.20: LSD results (marks) in 2D for a curved front system at when compared with the full model (solid): a) reconstructed enthalpy evolution in time. b) reconstructed sample temperature evolution in time. c) reconstructed Kirchhoff variable evolution in time. d) front evolution in time.
7.4. Model reduction

Therefore, an operator \( P_d \in \mathbb{R}^{d \times N} \) is needed to define the vector of measurements \( H_d = P_d H \), being \( H \) an enthalpy field vector such that \( H \in \mathbb{R}^N \). Note since the measurable variable is temperature, the measurements data must be firstly transformed into the corresponding enthalpy values.

The gain \( G \) is calculated according to the procedure also presented in Alonso et al. (2004) (code employed is presented in Appendix E). Hence, by adding and subtracting the term \( \Lambda m_H \) to the system in terms of the extensive variables modes, one gets:

\[
\frac{d\hat{m}_H}{dt} = -\Lambda m_E + \Lambda m_H - \Lambda m_H = \Lambda \hat{m}_H + f(\hat{m}_H)
\]

with \( f(\hat{m}_H) = \Lambda(\hat{m}_H - \hat{m}_E) \), being \( \hat{m}_H \) and \( \hat{m}_E \) the estimated temporal modes for the extensive variables, and \( \Lambda \) the diagonal matrix containing the eigenvalues obtained through LSD.

Then, the enthalpy modes can be reconstructed at a dynamic level from the corresponding measurements \( H_m \), by solving (Alonso et al., 2004):

\[
\frac{d\hat{m}_H}{dt} = \Lambda (\hat{m}_H - \hat{m}_E) + f(\hat{m}_H) + G(H_d - Q^T \hat{m}_H)
\]

being the matrix \( Q \) defined as follows:

\[
Q = \Phi^T P_d^T
\]

The gain matrix \( G \) determines the temporal rate at which the estimate modes converge to the real ones. The error associated to the observer is defined as:

\[
e_{obs} = m_H - \hat{m}_H
\]

Once the estimated modes for the enthalpy have been obtained, the enthalpy field \( H(x,t) \) can be reconstructed as follows:

\[
\hat{H} = \Phi \hat{m}_H
\]

and from this reconstructed enthalpy, the distributed product temperature can be also recovered by means of the relationships between extensive and intensive variables.

The numerical implementation of the observer system comprises both mechanisms
of on-line estimation and plant simulation. The latter one is intended to provide the required measurements. In this case, a homogeneous mesh of \( N = 101 \) nodes has been used, while the FEM method has been employed in order to solve the plant system and provide with \( d = 21 \) measurements to the observer layer. Besides, the LSD scheme has been made use of \( \text{neig} = 30 \) modes for reconstruction, but only \( \text{neig}_{\text{obs}} = 10 \) are observed. In addition, it has been assumed that the initial states of the system are unknown, which has been implemented by employing initial conditions far enough from the real ones.

The evolution of the two first estimated enthalpy modes is depicted in Figure 7.21 in comparison with the modes corresponding to the real system. Note the observer estimation convergence to the real values of the system, even though the difference on the initial conditions before mentioned. It is worthy to mention also that the convergence rate can be improved by employing a higher gain \( G \).

The results expressed in terms of the recovered distributed temperature field are shown in Figure 7.22 alongside the temperature distribution of the real system. The closest ones to the real temperature values coincides with those times for which the observer have already converged. Since the smoothing approach has been avoided, the effects of the Gibbs phenomenon are evident in the form of the oscillation around the phase change temperature value. However, the results are in a good agreement with the temperature of the full model used as reference.

Figure 7.21: Evolution of the two first estimated enthalpy modes compared to the evolution of the real system modes.
Figure 7.22: Recovered temperature field by employing the reduced-order observer scheme together with $d=21$ measurements: a) initial time. b) $t_{final}/4$ c) $t_{final}/2$ d) final time. The results are compared with the real system temperature obtained by simulation.

### 7.4.4 Template-fitting technique: off-line implementation

As previously commented in Chapter 3, similar characteristics can be found when comparing travelling waves to the Enthalpy-Kirchhoff transformed system. Based on this, a first approach to the application of the template-fitting, technique which has been described in Appendix D, is here presented.

This approach consists of the off-line implementation of the method. Therefore, the results of the one-dimensional model presented in Section 7.2.1 will be employed as data. For this particular example the following boundary conditions are imposed:

\[
T_I(0,t) = T_c > T_{change} \quad (7.38)
\]

\[
T_{II}(L,t) = T_L < T_{change} \quad (7.39)
\]

In essence, the template-fitting is applied to remove the travelling nature of the data in a given system by shifting the solution at all times to a chosen reference: the
so-called template (Rowley and Marsden, 2000). This could result really interesting for the case here studied, since it will simplified the computation of the reduced order model. The proposed scheme is outlined next.

- **Preliminaries.** A new working domain $[-L/2, 3L/2]$ obtained by extending the original one and padding with the boundary conditions value must be defined. Regarding the boundary conditions, the template-fitting technique demands them to be periodic. Therefore, the Enthalpy-Kirchhoff system will be again transformed in order to obtain homogeneous conditions, since by imposing $\bar{H}(0, t) = \bar{H}(L, t) = 0$ the periodicity requirement will be fulfilled. In this case, this transformation will be defined as:

$$\bar{H}(x,t) = H(x,t) - \frac{H_L - H_c}{L} x + H_c$$  \hfill (7.40)

- **Shifting.** To this purpose, the template must be firstly chosen. The reference considered will be the solution at time such that the front position is located at $x = L/2$, which will be noted as $H^\text{temp}$. Following the strategy presented in Runborg et al. (2002), the properties of the Fast Fourier Transform (FFT) will be employed to pin the system data to the chosen template. This new shifted data ensemble will be defined as follows:

$$\bar{H}(x + c(t), t) = \bar{H}(y, t)$$  \hfill (7.41)

with $y = x + c(t)$ and $c(t)$ being the shifting operator, which for this off-line application will be defined as the distance between the last dried node in the template and the last dried node position along the time. The shifting operator represents the displacement amount that each solution in the data ensemble requires to match the template.

- **ROM.** In accordance to the motivation behind the development of the template fitting technique, the Proper Orthogonal Decomposition (POD) will be employed in order to obtain the desired reduced representation of the shifted system.

- **Unshifting.** The Shifting theorem of the FFT is applied again to undo the data shifting. The original domain of the sample is recovered by removing the padding at the domain boundaries.
7.4. Model reduction

Figure 7.23: a) Enthalpy field with homogeneous boundary conditions after padding. b) Shifted enthalpy field with homogeneous boundary conditions.

Figure 7.23 shows the original enthalpy data and its corresponding shifted representation when considering homogeneous boundary conditions. The reduced solution obtained by employing only 41 POD basis (from the 101 equations solved in the full system) is presented in Figure 7.24, where it is has been compared to the full model (both depicted in the original domain). Please note the Gibbs phenomenon is not noticeable for this case.

Figure 7.24: Comparison between the full model (solid) and the ROM solution (marks): original domain when homogeneous boundary conditions are considered.
7.5 Conclusions

In this Chapter, the Enthalpy-Kirchhoff based method has been applied to two different Stefan-problems. The first of them consisted of a generic phase problem, where all the capabilities of this technique have been exposed. Both one-dimensional and two-dimensional scenarios have been solved, including a case exhibiting an irregular phase-change front. The second problem tackled, which corresponds to a real industrial problem (the freeze-drying of a dairy product) have revealed the difficulties associated to the method when the system configuration do not allows one-to-one mappings from intensive to extensive variables. However, an accurate algorithm to solve the problem in $1D$ and $2D$ without considering front curvature has been presented.

In addition, model reduction has been also applied to the generic Stefan problem considered. The Laplacian Spectral Decomposition technique has been chosen to obtain the low-dimensional representation of the system. Although the effects of the Gibbs phenomenon, in the shape of spurious oscillations have been revealed, different solutions to minimise its influence on the results obtained have been provided.

A smoothing approach has been firstly proposed, in combination with a filtering technique, in order to provide a continuous approximation of the enthalpy field. A reduction up to the 50% has been achieved, and a good agreement between reduced and full smooth systems has been obtained. However, the accuracy of these results with respect to the real system without smoothing has been revealed to be dependent on the interval $\Delta T$ employed to approximate the energy jump.

In order to overcome this drawback, a reduced-order observer scheme, which does not imply smooth extensive variables, has been developed. An on-line estimation scheme, in combination with the LSD-based low dimensional representation of the system has been employed for field reconstruction by means of a limited number of measurements of the system. Despite of the Gibbs phenomenon, results obtained showed a good agreement with respect to the real system combined with an important dimensional reduction (up to 60%).

As shown, the Enthalpy-Kirchhoff method constitutes an attractive method for modelling Stefan problems for systems exhibiting the suitable configuration (i.e monotonicity of variables). However, some difficulties need to be solved yet. In this regard, this work opened some alternatives to be explored. Future work will be focus on further exploitation of the Enthalpy-Kirchhoff system features. Due to the resemblance of the system with a travelling wave, the application of the template-fitting technique
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could represent an interesting option to be analysed, as the results of the off-line implementation have revealed.
The work presented in this PhD dissertation has been focus on the development of suitable operational models for real time applications of process distributed systems, with particular attention to moving boundary problems of interest in Food and Biotechnology industries.

The term operational model has been employed here in the sense of a mathematical model, based on first principles, whose derivation is carried out by considering both physical simplifications (relying on the insights of the system) and mathematical model reduction techniques. This modelling methodology results into a low dimensional and accurate representation of the system, through which the computational times employed for the numerical solving could have been significantly reduced. This becomes a key feature when jobs such as optimal control or on-line parameter estimation are tackled.

General concepts regarding modelling techniques of process distributed systems have been summarised in Part I of this dissertation. Classical techniques of solving PDE systems, such as the Finite Element Method (FEM) or the Finite Difference Method (FDM) have been outlined alongside those other alternative methods employed for model reduction. Among these latter ones, the Proper Orthogonal Decomposition (POD) and the Laplacian Spectral Decomposition (LSD) have been exposed.

Likewise, Part II has been devoted to illustrate the advantages of the proposed real-time oriented modelling scheme by means of two specific work cases.

The first of them exemplifies the developing of an operational model in the particular case of a compact heat exchange reactor. Drawbacks arise in relation to the highly exothermic reaction considered and the difficulty of maintaining reactor temperature
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in between the safety operation values.

- An operational model for the so called Open Plate Reactor has been derived and presented. Its performance has been validated by employing existing works in literature. In general, the model adequately reproduces the expected system behaviour. However, divergences with respect to literature have been found and reported.

- Two different model reduction techniques have been applied in order to find the low-order representation of the OPR system: the Laplacian Spectral Decomposition (LSD) and the Proper Orthogonal Decomposition (POD). It has been found that the reduction power of the LSD method is strongly determined by the characteristic Péclet number of the reactor. Comparison between systems exhibiting low and high Péclet values has been offered, showing that systems in which diffusive mechanism is important, namely low Péclet number systems, are more adequate for the LSD technique.

In addition, the POD method has been revealed as an effective method to obtain the desired reduced model for the OPR, achieving a reduction up to the 50% of the computation time with respect to the FEM solved system. Besides, POD based dimensional reduction is efficient independently of the diffusive phenomenon relevance (i.e. for all the Péclet numbers values).

- The POD-based low dimensional representation of the system has been employed as the core of the optimal control strategy presented. Two different scenarios have been devised in order to achieve the desired objective: to start-up the OPR operation satisfying the safety constraints while maximising the reactants conversion. The first case consisted of a DO without restrictions, while the second one introduced constraints on the state variables. The utilisation of this ROM scheme has clearly decreased the computational times (around 1000 evaluations of the objective function are carried out in 30 seconds), while eased the system analysis.

On the other hand, the second work case is focused on the application of the proposed modelling scheme to moving boundary problems, specifically those problems exhibiting a moving front associated to a phase-change, namely Stefan problems.

- Conceptual modelling have been revealed as very useful tool to achieve a better understanding of freeze-drying process dynamics and their influence on cycle

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time and product temperature history, which are considered to be the key parameters for product quality, stability and productivity.

A first principle based model has been derived from a rigorous time-scale analysis, which has made use of the inherent thermophysical properties of the material, in order to define the different time scales involved in the operation.

The time-scale reduction approach has enabled to focus just on the phenomenon of interest, namely the time dependent product temperature distribution, and to neglect mechanism occurring at faster scales. Thus a simplified a low-dimensional control-oriented model has been obtained, being its performance validated by comparison with the existent literature.

- The matrix scale model has been employed as the core of the proposed optimal control approach, which defines the operation conditions for minimizing freeze-drying cycle time while preserving product quality (final water content) through the solution of a dynamic NLP.

Diverse control scenarios have been analysed. First, constant optimal control profiles for $T_L$ and $P_c$ along the process time horizon were considered, overcoming temperature constraint violations that could lead to collapse of the product porous structure. For this scenario, final product quality is ensured at the expenses of an increase of about the 29% of the cycle time as compared with a standard non-optimal constant profile ($T_L = 263$ $K$ and $P_c = 10$ $Pa$).

After that, variable control profiles were proved to be successful in reducing process time while satisfying product quality standards. The resulting optimal profiles for $(T_L, P_c)$ led to reductions of up to an 17.71% (one control variable case - $T_L$) and 25.5% (two control variable case - $T_L$ and $P_c$) of the cycle time when compared with the case under optimal constant controls ($T_L = 251.75$ $K$ and $P_c = 25.398$ $Pa$).

- A new non-invasive methodology for estimating on-line the product temperature distribution during the primary-drying of freeze-drying processes has been presented. This methodology also provides fair estimations for the front temperature and position on a three-steps scheme basis:

  1. The so called Pressure Rise Tests (PRT) have been employed to obtain reliable estimates for the sublimation front temperature.
2. The Landau transform have been applied to the original moving boundary problem to obtain an equivalent system representation with fixed spatial domain. The estimates provided by the PRT step are employed here to obtain the closure of the PDE system formulated in the new fixed space.

3. Model reduction by means of the application of the Proper Orthogonal Decomposition (POD) technique has been employed to decrease the dimensionality and lighten the computational costs of the transformed system.

Simulation experiments were carried out, and their results were compared to the accurate numerical solutions provided a commercial FEM software (COMSOL Multyphysics©) which employs an ALE method to solve the moving front problem. The estimates obtained are in good agreement with the COMSOL solutions. Slight differences can be appreciated at the final part of the process \( t > 30 \text{h} \) because the time required to reach equilibrium increases in this period as a result of the vapour flux drop.

- The Enthalpy-Kirchhoff based method has been applied to two different Stefan-problems. The first of them consisted of a generic phase problem, where all the capabilities of this techniques have been exposed. Both one-dimensional and two-dimensional scenarios have been solved, including a case exhibiting an irregular phase-change front.

The second problem tackled, which corresponds to a real industrial problem (the freeze-drying of a dairy product), have revealed the difficulties associated to the method when the system configuration do not allows one-to-one mappings from intensive to extensive variables. However, an accurate algorithm to solve the problem in 1D and 2D without considering front curvature has been presented.

In addition, model reduction has been also applied to the first scenario considered. The Laplacian Spectral Decomposition technique has been chosen to obtain the low-dimensional representation of the system. Although the effects of the Gibbs phenomenon have been revealed in the shape of spurious oscillations, different solutions to minimise its influence on the results obtained have been provided.

A smoothing approach has been firstly proposed, in combination with a filtering technique, in order to provide a continuous approximation of the enthalpy field.
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A reduction up to the 50% has been achieved, and a good agreement between reduced and full smooth systems has been obtained. However, the accuracy of these results with respect to the real system without smoothing has been revealed to be dependent on the interval $\Delta T$ employed to approximate the energy jump.

In order to overcome this drawback, a reduced-order observer scheme, which not implies smooth extensive variables, has been developed. An on-line estimation scheme, in combination with the LSD-based low dimensional representation of the system has been employed for field reconstruction by means of a limited number of measurements of the system. Despite of the Gibbs phenomenon, results obtained showed a good agreement with respect to the real system combined with an important dimensional reduction (up to 60%).

As shown, the Enthalpy-Kirchhoff method constitutes an attractive method for modelling Stefan problems for systems exhibiting the suitable configuration (i.e monotonicity of variables). However, some difficulties need to be solved yet. In this regard, this work opened some alternatives to be explored. Future work will be focus on further exploitation of the Enthalpy-Kirchhoff system features. Due to the resemblance of the system with a travelling wave, the application of the template-fitting technique could represent an interesting option to be analysed, as the results of the off-line implementation have revealed.
Conclusións

O traballo presentado nesta tese está centrado no desenvolvemento de modelos operacionais axeitados para aplicacións en tempo real de procesos distribuídos, con particular énfase en problemas de fronteira móbil de especial interese nas industrias alimentaria e biotecnolóxica.

O concepto de *modelo operacional* responde a aqueles modelos matemáticos, baseados en primeiros principios, que se derivan de considerar simplificacións físicas (apoiadas no estudo e comprensión do sistema) e técnicas matemáticas de redución de modelos. Esta metodoloxía de modelaxe achega unha representación de baixa dimensionalidade e precisa do sistema, grazas á que é posible reducir significativamente os tempos computacionais empregados na súa resolución numérica. Esta característica resulta clave cando se abordan problemas de control óptimo ou estimación en liña de parámetros.

Os conceptos xerais relacionados coas técnicas de modelaxe de procesos de sistemas distribuídos poden atoparse na Parte I desta tese. As técnicas clásicas de resolución de sistemas de Ecuacións en Derivadas Parciais (EDP), tales coma o Método de Elementos Finitos (MEF) ou o Método das Diferenzas Finitas (MDF) aparecen aí descritos xunto con outros métodos empregados como alternativa e encamiñados á redución de modelos. Entre estes últimos atópanse a Descomposición Ortogonal Propia (POD nas súas siglas inglesas) e a Descomposición Espectral do Laplaciano (LSD).

Do mesmo xeito, a Parte II está adicada a ilustrar, por medio de dous casos específicos, as vantaxes do esquema de modelaxe orientado a aplicacións en tempo real.
Conclusións

O primeiro destes casos exemplifica o desenvolvemento dun modelo operacional para o caso particular dun reactor intercambiador de calor de tipo compacto. Os inconvenientes xorden en relación á reacción altamente exotérmica considerada e a dificultade que supón manter a temperatura do reactor entre os valores de operación de marcados polas normas de seguridade.

- Derivouse un modelo operacional para o denominado Open Plate Reactor. O seu comportamento foi validado a través do emprego por comparación con traballos previos existentes na bibliografía. En xeral, este modelo proposto reproduce adecuadamente o comportamento esperado do sistema. Con todo, atopáronse certas diferenzas con respecto a resultados xa existentes que foron pertinentemente comentadas.

- Dúas técnicas diferentes de redución de modelos foron aplicadas a fin de obter unha representación de baixa dimensionalidade para o sistema representativo do reactor OPR: a Descomposición Espectral do Laplaciano e a Descomposición Ortogonal Propia.

Atopouse que o poder de redución da Descomposición Espectral do Laplaciano está fortemente determinado polo valor do Péclet característico do reactor. Comparáronse dous sistemas con distinto número Péclet, un deles alto e o outro baixo. Os resultados obtidos amosaron que aqueles sistemas nos que o mecanismo de difusión é máis importante, isto é, os que teñen Péclet baixo, son os máis adecuados para a aplicación da técnica de Descomposición Espectral do Laplaciano.

Ademais, o método da Descomposición Ortogonal Propia revelouse como unha ferramenta efectiva á hora de obter o desexado modelo de orde reducida para o reactor OPR, acadándose unha diminución de ata o 50% no tempo computacional empregado para resolver o modelo en comparación co empregado ó utilizar o Método dos Elementos Finitos. Tamén se puxo de manifiesto que a redución de modelos baseada no emprego da Descomposición Ortogonal Propia e totalmente independente da importancia do fenómeno difusivo (isto é, independente dos valores de Péclet).

- A representación de baixa dimensionalidade obtida mediante Descomposición Ortogonal Propia foi empregada como núcleo da estratexia de control óptimo
presentada. Trazáronse dous escenarios diferentes para tentar acadar o obxectivo seguinte: realizar unha posta en marcha do reactor satisfacendo os requisitos de seguridade ó tempo que se maximizaba a conversión de reactivos.

O primeiro caso proposto consistiu nun problema de optimización dinámica sen restricións, namente que no segundo se introduciron restricións sobre as variables de estado. A utilización do esquema de orde reducida permitiu diminuír considerablemente o tempo computacional (unhas 1000 avaliacións da función obxectivo realizápanse en 30 segundos), ó tempo que facilitou a análise do sistema.

Por outra banda, o segundo caso de traballo centrouse na aplicación do esquema de modelaxe proposto a problemas de fronteira móbil, específicamente aqueles que involucran unha fronte en movemento asociada a un cambio de fase: os denominados problemas de Stefan.

- Este tipo de modelaxe conceptual revelouse como unha ferramenta moi conveniente para acadar un mellor entendemento da dinámica do proceso de liofilización de alimentos e da súa influencia no tempo do ciclo e no histórico de temperaturas do sistema, que son considerados os parámetros clave que determinan a calidade do produto, a súa estabilidade e a súa produtividade.

Dixese un modelo baseado en primeiros principios a partir das dunha rigorosa análise das escalas de tempo. Dita análise fixo uso das propiedades termofísicas inherentes do material co obxectivo de definires as escalas de tempo involucradas na operación de liofilización.

Esta reducción baseada nas escalas de tempo fixo posible ter en conta soamente o fenómeno de interese, isto é a distribución de temperatura no produto ó longo do tempo, e obviar outros mecanismos que ocorrían a diferentes escalas. Obtívose así un modelo de baixa dimensionalidade e orientado ó control en tempo real, que foi validado empregando datos da bibliografía existente.

- O modelo na escala de tempo da matriz porosa foi empregado como núcleo da estratexia de control óptimo proposta, que definiu as condicións óptimas de operación para minimizar o tempo do proceso liofilización garantindo a calidade do produto (contido final de auga) mediante a resolución dun problema NLP dinámico.

Analizáronse distintos escenarios de control. Primeiramente, consideráronse perfís constantes para as variables de control $T_L$ e $P_L$ ó longo de todo o horizonte
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de tempo do proceso, observándose violaciones nas restriccions impostas sobre a temperatura que poderían levar ó colapso da estrutura porosa do produto. Para este escenario, a calidade final do produto asegurouse a expensas dun incremento do 29% no tempo de ciclo en comparación co perfil constante non óptimo de referencia ($T_L = 263$ K e $P_c = 10$ Pa).

A continuación, ensaiáronse perfís de control variables no tempo, que resultaron ser adecuados para reducir o tempo do proceso namentres que se mantiña a calidade do produto nos niveis requeridos polo mercado. Os resultantes perfís de control óptimos para ($T_L, P_c$) permitiron reducir un 17.71% (caso con unha variable de control - $T_L$) e un 25.5% (caso con dúas variables de control - $T_L$ e $P_c$) o tempo dun ciclo de operación en comparación co caso definido por controles constantes ($T_L = 251.75$ K e $P_c = 25.398$ Pa).

• Presentouse unha novidosa metodoloxía deseñada para estimar en liña a distribución de temperatura do produto durante o secado primario do proceso de liofilización. Esta metodoloxía tamén proporcionou estimacións fiables da temperatura da fronte de sublimación e da súa posición mediante un esquema de tres pasos:


2. A transformación de Landau foi tamén aplicada ó problema orixinial de fronteira móbil para conseguir unha representación equivalente do sistema nun dominio espacial fixo. As estimacións proporcionadas polo paso anterior do esquema (PRT) utilizáronse aquí para pechar o sistema en derivadas parciais formulado no novo espacio de traballo.

3. Redución do modelo mediante Descomposición Ortogonal Propia, que permitiu diminuír a dimensionalidade do sistema e reduciu os custos computacionais asociados ó novo sistema transformado.

Leváronse a cabo experimentos de simulación, e os seus resultados foron comparados coas solucións obtidas para o sistema completo mediante un software comercial de Elementos Finitos (COMSOL Multyphysics®), no que se emprega un algoritmo ALE na resolución do problema de fronteira móbil. As estimacións obtidas amosáronse en boa concordancia cos resultados correspondentes ó sistema completo. Sen embargo, detectáronse lixeiras diferencias na parte final.
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do proceso \((t > 30\, \text{h})\), debido a que o tempo requirido para acadar o equilibrio se ve aumentado a consecuencia da caída no fluxo de vapor nesta etapa do proceso.

- Aplicouse o método baseado na transformación en Entalpía e Kirchhoff a dous problemas de Stefan distintos. O primeiro deles consistiu nun problema de cambio de fase xenérico, onde as capacidades desta técnica foron postas de manifiesto. Resolvéronse tanto o problema unidimensional como o correspondente problema en dúas dimensións, incluíndo un caso no que a fronte do cambio de fase presentaba unha forma irregular.

O segundo problema tratado, correspondente a un problema industrial real (a liofilización dun produto lácteo), serviu para poñer de manifiesto as dificultades asociadas a este método cando a configuración do sistema non permite correspondencias un a un entre as variables intensivas e as variables extensivas. Aínda así, conseguiuse desenvolver un algoritmo para resolver problemas en \(1D\) e \(2D\) para aqueles caso nos que non se considere a curvatura da fronte.

Ademais, obtívose a correspondente representación de orde reducida para o primeiro escenario presentado (problema xenérico). A Descomposición Espectral do Laplaciano foi a técnica empregada neste caso. A pesares de que os efectos do fenómeno de Gibbs se manifestaron en forma de oscilacións espúreas, presentáronse diversas estratexias encamiñadas a minimizar a súa influencia nos resultados obtidos.

A primeira destas estratexias baseouse nunha aproximación \textit{smooth} da variable discontinua en combinación con unha técnica de filtrado. Obtívose unha redución do arredor do 50% no número de graos de liberdade do sistema, estando o modelo reducido en boa concordancia co comportamento do modelo completo. Sen embargo, atopouse que a exactitude destes resultados con respecto ó modelo real (sen \textit{smoothing}) depende do intervalo \(\Delta T\) empregado para aproximar o salto de enerxía na variable entalpía.

Co obxectivo de resolver esta dificultade, desenvolveuse un observador baseado no modelo reducido do sistema que non require da aproximación \textit{smoothing} das variables extensivas. Empregouse un esquema de estimación en liña, en combinación co modelo de orde reducida obtido mediante LSD, para a reconstrución da variable entalpía mediante un número limitado de medidas experimentais.
Apesares do fenómeno de Gibbs, os resultados obtidos amosaron unha boa correspondencia co sistema real, ó tempo que se acadou unha importante redución do sistema (aproximadamente do 60%).

Finalmente, tamén se presentou un sinxelo exemplo que debe ser tomado como paso previo na aplicación da técnica do template-fitting para a formulación en variables extensivas dos problemas de Stefan.
Part III

Appendixes
A.1 Two-dimensional freezing case

Let be $\gamma(x, y, t) = y - S(x, t) = 0$, with the moving front position defined by $y = S(x, t)$. The general form for the Stefan condition used on Equation (3.17) is now developed for the two-dimensional case term by term. The starting point will be the general energy balance across the moving boundary:

$$[\rho e]_s^n v_n = [\overrightarrow{q}]_s^n n$$  \hspace{1cm} (A.1)

First term

First of all, the definition of the velocity and the normal vector to $\gamma(x, y, t) = 0$ in 2D is presented:

$$\overrightarrow{v} = (v_x, v_y) = \left( \frac{dx}{dt}, \frac{dy}{dt} \right)$$  \hspace{1cm} (A.2)

$$\overrightarrow{n} = \frac{\overrightarrow{\nabla} \gamma}{|\overrightarrow{\nabla} \gamma|} = \left( \frac{\partial \gamma}{\partial x}, \frac{\partial \gamma}{\partial y} \right) \frac{1}{|\overrightarrow{\nabla} \gamma|}$$  \hspace{1cm} (A.3)

The normal velocity for any point of the moving front $\gamma(x, y, t) = 0$ is given by the following scalar product:

$$v_n = \overrightarrow{v} \cdot \overrightarrow{n} = \left( \frac{dx}{dt}, \frac{dy}{dt} \right) \left( \frac{\partial \gamma}{\partial x}, \frac{\partial \gamma}{\partial y} \right) \frac{1}{|\overrightarrow{\nabla} \gamma|} = \left( \frac{\partial \gamma}{\partial x} \frac{dx}{dt} + \frac{\partial \gamma}{\partial y} \frac{dy}{dt} \right) \frac{1}{|\overrightarrow{\nabla} \gamma|}$$  \hspace{1cm} (A.4)

The time derivative of $\gamma(x, y, t)$ can be written as:

$$\frac{d\gamma}{dt} = \frac{\partial \gamma}{\partial x} \frac{dx}{dt} + \frac{\partial \gamma}{\partial y} \frac{dy}{dt} + \frac{\partial \gamma}{\partial t} = 0$$  \hspace{1cm} (A.5)
and therefore:

\[-\frac{\partial \gamma}{\partial t} = \frac{\partial \gamma}{\partial x} \frac{dx}{dt} + \frac{\partial \gamma}{\partial y} \frac{dy}{dt}\]  \hspace{1cm} (A.6)

So, by substituting Eqn (A.6) into Eqn (A.4) the normal velocity can be redefined by:

\[-\vec{v} \cdot \vec{n} = - \frac{\partial \gamma}{\partial t} \frac{1}{|\vec{\nabla} \gamma|}\]  \hspace{1cm} (A.7)

Time differentiation of $\gamma(x, y, t) = y - S(x, t)$ results into:

\[-\frac{\partial \gamma}{\partial t} = - \frac{\partial S(x, t)}{\partial t}\]  \hspace{1cm} (A.8)

which in combination with Eqn (A.8) leads to the general expression for the first term in Eqn (A.1):

\[\left[\rho e\right]_s l v_n = \rho_s \Delta H \frac{\partial S(x, t)}{\partial t} \frac{1}{|\vec{\nabla} \gamma|}\]  \hspace{1cm} (A.9)

Second term

The second term involves the Fourier’s law, so let’s define the temperature gradient as:

\[\vec{\nabla} T = \left(\frac{\partial T}{\partial x}, \frac{\partial T}{\partial y}\right)\]  \hspace{1cm} (A.10)

At $\gamma(x, y, t) = 0$, a continuity condition for temperature is also imposed:

\[T_s\big|_{\gamma=0} = T_l\big|_{\gamma=0} = T_f\]  \hspace{1cm} (A.11)

where $T_f$ is the constant freezing temperature. By differentiation of $T(x, y, t)$ respect to independent variable $x$:

\[\frac{dT}{dx} = \frac{\partial T}{\partial y} \frac{dy}{dx} + \frac{\partial T}{\partial x} = 0\]  \hspace{1cm} (A.12)

which after rearranging terms can be expressed as:

\[-\frac{\partial T}{\partial x} = - \frac{\partial T}{\partial y} \frac{dy}{dx}\]  \hspace{1cm} (A.13)

Then, taking into account that:

\[\frac{d\gamma}{dx} = \frac{\partial \gamma}{\partial y} \frac{dy}{dx} + \frac{\partial \gamma}{\partial x} = 0\]  \hspace{1cm} (A.14)
A.2. Three-dimensional freezing case

leads to:
\[
\frac{dy}{dx} = \frac{-\frac{\partial \gamma}{\partial x}}{\frac{\partial \gamma}{\partial y}} \tag{A.15}
\]

Considering both Eqn (A.13) and Eqn (A.15), it is possible to rewrite Eqn (A.10) as follows:
\[
\nabla T = \left( -\frac{\partial T}{\partial y} \frac{dy}{dx}, \frac{\partial T}{\partial y} \right) = \frac{\partial T}{\partial y} \left( -\frac{\partial S(x,t)}{\partial x}, 1 \right) \tag{A.16}
\]

Recovering the general expression for the second term, and using the above equations:
\[
\left[ \vec{q}' \right]_{n} = \left[ \left( -k_{l} \frac{\partial T_{l}}{\partial y} + k_{s} \frac{\partial T_{s}}{\partial y} \right) \right] \left[ \left( -\frac{\partial S(x,t)}{\partial x}, 1 \right) \right] \frac{1}{|\nabla \phi|} =
\]
\[
\left[ \left( -k_{l} \frac{\partial T_{l}}{\partial y} \bigg|_{S(x,y,t)^{+}} + k_{s} \frac{\partial T_{s}}{\partial y} \bigg|_{S(x,y,t)^{-}} \right) \right] \left[ \left( \frac{\partial S(x,t)}{\partial x} \right)^{2} + 1 \right] \frac{1}{|\nabla \gamma|} \tag{A.17}
\]

Finally, Eqn (A.17) together with (A.9) lead to the final expression for the Stefan condition in 2D, which reads:
\[
\frac{\partial S(x,t)}{\partial t} \Delta H \rho_{s} = \left[ -k_{l} \frac{\partial T_{l}}{\partial y} \bigg|_{S(x,y,t)^{+}} + k_{s} \frac{\partial T_{s}}{\partial y} \bigg|_{S(x,y,t)^{-}} \right] \left[ \left( \frac{\partial S(x,t)}{\partial x} \right)^{2} + 1 \right] \tag{A.18}
\]

### A.2 Three-dimensional freezing case

Let be now function \( \gamma(x, y, z, t) = z - S(x, y, t) = 0 \), where \( S(x, y, t) \) represents the position of the moving front.

When considering a 3D case, the gradient of function \( \gamma(x, y, z, t) \) will be:
\[
\nabla \gamma = \left( \frac{\partial \gamma}{\partial x}, \frac{\partial \gamma}{\partial y}, \frac{\partial \gamma}{\partial z} \right) \tag{A.19}
\]

with
\[
\frac{\partial \gamma}{\partial x} = -\frac{\partial S(x,y,t)}{\partial x}, \quad \frac{\partial \gamma}{\partial y} = -\frac{\partial S(x,y,t)}{\partial y}, \quad \frac{\partial \gamma}{\partial z} = 1 \quad \tag{A.20}
\]

and therefore:
\[
\nabla \gamma = \left( \frac{\partial \gamma}{\partial x}, \frac{\partial \gamma}{\partial y}, \frac{\partial \gamma}{\partial z} \right) = \left( -\frac{\partial S(x,y,t)}{\partial x}, -\frac{\partial S(x,y,t)}{\partial y}, 1 \right) \tag{A.21}
\]
The normal vector to $\gamma(x, y, z, t) = 0$ is defined as

$$\vec{n} = \frac{\vec{\nabla} \gamma}{|\vec{\nabla} \gamma|}$$  \hspace{1cm} (A.22)

and the velocity vector of a point in the phase change front for this three dimensional case will read:

$$\vec{v} = (v_x, v_y, v_z) = \left(\frac{dx}{dt}, \frac{dy}{dt}, \frac{dz}{dt}\right)$$  \hspace{1cm} (A.23)

By differentiation of $\gamma(x, y, z, t) = z - S(x, y, t) = 0$ ones gets:

$$\frac{d\gamma}{dt} = \frac{\partial \gamma}{\partial x} \frac{dx}{dt} + \frac{\partial \gamma}{\partial y} \frac{dy}{dt} + \frac{\partial \gamma}{\partial z} \frac{dz}{dt} + \frac{\partial \gamma}{\partial t} = 0$$  \hspace{1cm} (A.24)

which after rearranging terms leads to:

$$- \frac{\partial \gamma}{\partial t} = \frac{\partial \gamma}{\partial x} \frac{dx}{dt} + \frac{\partial \gamma}{\partial y} \frac{dy}{dt} + \frac{\partial \gamma}{\partial z} \frac{dz}{dt}$$  \hspace{1cm} (A.25)

By combining Eqn (A.22) and Eqn (A.23) with Eqn (A.25), it is obtained:

$$\vec{v} \cdot \vec{n} = - \frac{\partial \gamma}{\partial t} \frac{1}{|\vec{\nabla} \gamma|}$$  \hspace{1cm} (A.26)

Thus, the first term of the Stefan condition for the 3D case can be rewritten as:

$$[\rho_e] v_n = \rho_s \Delta H \frac{\partial S(x, y, t)}{\partial t} \frac{1}{|\vec{\nabla} \gamma|}$$  \hspace{1cm} (A.27)

**Second term**

For a 3D case, the temperature gradient reads:

$$\vec{\nabla} T = \left(\frac{\partial T}{\partial x}, \frac{\partial T}{\partial y}, \frac{\partial T}{\partial z}\right)$$  \hspace{1cm} (A.28)

Considering the isotherm condition imposed at the freezing front $T_s = T_l = T_f$, and by differentiation of $T(x, y, z)$:

$$\frac{\partial T}{\partial x} = - \frac{\partial T}{\partial z} \frac{dz}{dx}; \quad \frac{\partial T}{\partial y} = - \frac{\partial T}{\partial z} \frac{dz}{dy}$$  \hspace{1cm} (A.29)
A.2. Three-dimensional freezing case

Equation (A.28) can be rewritten as

\[ \vec{\nabla} T = \left( -\frac{\partial T}{\partial z} \frac{dz}{dx}, -\frac{\partial T}{\partial z} \frac{dz}{dy}, \frac{\partial T}{\partial z} \right) = \frac{\partial T}{\partial z} \left( \frac{dz}{dx}, \frac{dz}{dy}, 1 \right) \]  

(A.30)

Differentiation of \( \gamma(x, y, z, t) \) leads to:

\[ \frac{dz}{dx} = -\frac{\partial \gamma}{\partial x} \frac{\partial \gamma}{\partial z}; \quad \frac{dz}{dy} = -\frac{\partial \gamma}{\partial y} \frac{\partial \gamma}{\partial z} \]  

(A.31)

therefore, the gradient can be expressed as:

\[ \vec{\nabla} T = \frac{\partial T}{\partial z} \left( -\frac{\partial \gamma}{\partial x} \frac{\partial \gamma}{\partial z}, -\frac{\partial \gamma}{\partial y} \frac{\partial \gamma}{\partial z}, 1 \right) = \frac{\partial T}{\partial z} \left( -\frac{\partial S(x, y, t)}{\partial x}, -\frac{\partial S(x, y, t)}{\partial y}, 1 \right) \]  

(A.32)

(A.33)

Now, after recovering the Fourier law, and combine it with the above equations, the second term expression will be defined as follows:

\[ \left[ \begin{array}{c} \vec{q} \\ \vec{n} \end{array} \right] = \left[ \begin{array}{c} -k_l \frac{\partial T_l}{\partial z}\big|_{S(x,y,t)}^{+} + k_s \frac{\partial T_s}{\partial z}\big|_{S(x,y,t)}^{-} \\ \left( \frac{\partial S(x, y, t)}{\partial x} \right)^2 + \left( \frac{\partial S(x, y, t)}{\partial y} \right)^2 + 1 \end{array} \right] \frac{1}{|\vec{\nabla} \gamma|} \]  

(A.34)

(A.35)

Therefore, considering Eqn (A.27) and Eqn (A.34), the 3D Stefan condition will be written as:

\[ \rho_s \Delta H \frac{\partial S(x, y, t)}{\partial t} = \left[ \begin{array}{c} -k_l \frac{\partial T_l}{\partial z}\big|_{S(x,y,t)}^{+} + k_s \frac{\partial T_s}{\partial z}\big|_{S(x,y,t)}^{-} \\ \left( \frac{\partial S(x, y, t)}{\partial x} \right)^2 + \left( \frac{\partial S(x, y, t)}{\partial y} \right)^2 + 1 \end{array} \right] \]  

(A.36)

(A.37)
B.1 Application of the Landau transform to a rectangular domain

Here is presented the Landau transform applied to a cartesian 2D domain, where the function $\gamma(x,y,t) = y - S(x,t) = 0$ is considered again, representing $y = S(x,t)$ the moving front, which is supposed to evolve in the $Y$ axis. The spatial subdomains considered before:

$$\Omega_s = \{(x,y) \in \mathbb{R}^2 | 0 \leq x \leq D, 0 \leq y \leq S(x,t), t > 0\}$$
$$\Omega_l = \{(x,y) \in \mathbb{R}^2 | 0 \leq x \leq D, S(x,t) \leq y \leq L, t > 0\}$$

and after the transformation:

$$\tilde{\Omega}_s = \{(x,z_s) \in \mathbb{R}^2 | 0 \leq x \leq D, 0 \leq z_s \leq 1, t > 0\}$$
$$\tilde{\Omega}_l = \{(x,z_l) \in \mathbb{R}^2 | 0 \leq x \leq D, 0 \leq z_l \leq 1, t > 0\}$$

are represented in Figure B.1. The new spatial variables $z_s$ and $z_l$ introduced in order to fix the boundaries are:

$$z_s = \frac{y}{S(x,t)} = \begin{cases} 0 & \text{if } y = 0 \\ 1 & \text{if } y = S(x,t) \end{cases} \quad 0 \leq z_s \leq 1 \quad (B.1)$$

$$z_l = \frac{y - S(x,t)}{L - S(x,t)} = \begin{cases} 0 & \text{if } y = S(x,t) \\ 1 & \text{if } y = L \end{cases} \quad 0 \leq z_l \leq 1 \quad (B.2)$$
The original phase change problem in 2D is described by:

\[
\frac{\partial T_s}{\partial t} = \alpha_s \left( \frac{\partial^2 T_s}{\partial x^2} + \frac{\partial^2 T_s}{\partial y^2} \right) \quad \forall (x, y) \in \Omega_s \tag{B.3}
\]

\[
\frac{\partial T_l}{\partial t} = \alpha_l \left( \frac{\partial^2 T_l}{\partial x^2} + \frac{\partial^2 T_l}{\partial y^2} \right) \quad \forall (x, y) \in \Omega_l \tag{B.4}
\]

with external boundary conditions:

\[
T_s(0, y, t) = T_s(D, y, t) = T_w; \quad T_l(0, y, t) = T_l(D, y, t) = T_w \tag{B.5}
\]

\[
T_s(x, 0, t) = T_c; \quad T_l(x, L, t) = T_L
\]

At the moving front, the temperature continuity:

\[
T_s(x, S(x, t), t) = T_l(x, S(x, t), t) = T_f \tag{B.6}
\]

together with the Stefan condition are imposed:

\[
\rho_s \Delta D \frac{\partial S(x, t)}{\partial t} = \left( -k_l \frac{\partial T_l}{\partial y} + k_s \frac{\partial T_s}{\partial y} \right) \left[ \left( \frac{\partial S(x, t)}{\partial x} \right)^2 + 1 \right] \tag{B.7}
\]
After the coordinate transformation defined in Eqn (B.1) and Eqn (B.2), the original system becomes into:

\[
\frac{\partial \tilde{T}_l}{\partial \theta} - z_s S \frac{\partial \tilde{T}_s}{\partial z_s} = \alpha_s \left( \frac{\partial^2 \tilde{T}_s}{\partial x^2} + \frac{1}{S^2} \frac{\partial^2 \tilde{T}_s}{\partial z_s^2} \right) \quad \forall (x, z_s) \in \tilde{\Omega}_s
\]  
(B.8)

\[
\frac{\partial \tilde{T}_l}{\partial \theta} - S \left( 1 - z_l \right) \frac{\partial \tilde{T}_l}{\partial z_l} = \alpha_l \left( \frac{\partial^2 \tilde{T}_l}{\partial x^2} + \frac{1}{(L - S)^2} \frac{\partial^2 \tilde{T}_l}{\partial z_l^2} \right) \quad \forall (x, z_l) \in \tilde{\Omega}_l
\]  
(B.9)

where \( S \) notes \( S(x, \theta) \). The boundary conditions corresponding to the external boundaries will read:

\[
\tilde{T}_s(0, z_s, \theta) = \tilde{T}_s(D, z_s, \theta) = \tilde{T}_w; \quad \tilde{T}_l(0, z_l, \theta) = \tilde{T}_l(D, z_l, \theta) = \tilde{T}_w
\]  
(B.10)

\[
\tilde{T}_s(x, 0, \theta) = \tilde{T}_c; \quad \tilde{T}_l(x, 1, \theta) = \tilde{T}_L
\]

while those other ones imposed at the front are rewritten in the following form:

\[
\tilde{T}_s(x, 1, \theta) = \tilde{T}_l(x, 0, \theta) = \tilde{T}_f
\]  
(B.11)

\[
\rho_s \Delta D \frac{\partial S}{\partial \theta} = \left( \frac{k_s \tilde{T}_s}{S} \frac{\partial \tilde{T}_s}{\partial z_s} - \frac{k_l \tilde{T}_l}{(L - S)} \frac{\partial \tilde{T}_l}{\partial z_l} \right) \left[ \left( \frac{\partial S}{\partial x} \right)^2 + 1 \right]
\]  
(B.12)
C.1 Justification

Let’s consider again the Stefan problem formulated in Section 3.3.3:

\[
\frac{\partial H}{\partial t} = \frac{\partial^2 E}{\partial x^2}, \quad x \in (0, L) \quad (C.1)
\]

\[
E(0, t) = E_c; \quad \frac{\partial E}{\partial x}(L, t) = 0 \quad (C.2)
\]

\[
H(x, 0) = H(T(x, 0)) \quad (C.3)
\]

in the time range \(0 \leq t \leq t_f\). The first step will be to obtain the weak formulation of the above problem. It is assumed that higher-order derivatives of \(H(T)\) and \(E(T)\) exist, and that the test function \(\Psi(x, t) \in C^2(\Omega)\) at least, with \(\Psi(0, t) = \Psi(L, t) = 0\). Thus, considering Eqn \((C.1)\) in distributional sense:

\[
0 = \langle \frac{\partial H}{\partial t} - \frac{\partial^2 E}{\partial x^2}, \Psi \rangle \equiv \int_0^{t_f} \int_{\Omega} \left( \frac{\partial H}{\partial t} \Psi - \frac{\partial E}{\partial x} \frac{\partial^2 \Psi}{\partial x^2} \right) \, dx \, dt \quad (C.4)
\]

\[
\equiv \int_0^{t_f} \int_{\Omega} \left( -H \frac{\partial \Psi}{\partial t} + E \frac{\partial^2 \Psi}{\partial x^2} \right) \, dx \, dt
\]

and after integrating by parts, the weak formulation is obtained:

\[
\int_0^{t_f} \int_{\Omega} \left( H \frac{\partial \Psi}{\partial t} + E \frac{\partial^2 \Psi}{\partial x^2} \right) \, dx \, dt = \quad (C.5)
\]

\[
= -\int_{\Omega} H(T_{ini}) \Psi(x, 0) \, dx - \int_0^{t_f} \frac{\partial \Psi}{\partial x}(0, t) E(0, t) \, dt + \int_0^{t_f} \Psi(L, t) \frac{\partial E}{\partial x}(L, t) \, dt
\]
In order to show that the weak formulation includes the Stefan condition across the phase-change surface, let’s suppose now that the domain is divided into two different regions, one for each phase, as seen in Figure C.1. So, noting by $\Omega_s$ the solid phase by $\Omega_l$ the liquid material, and after integrating by parts on the two subdomains, one gets:

\[ \int_0^{t_f} \int_{\Omega_s} \left( H \frac{\partial \Psi}{\partial t} + E \frac{\partial^2 \Psi}{\partial x^2} \right) \, dx \, dt = \] (C.6)

\[ = - \int_0^{S(t=0)} H(T_{ini}) \Psi(x,0) \, dx - \int_0^{t_f} \frac{\partial \Psi}{\partial x}(0,t) E(0,t) \, dt + \] 

\[ + \int_{S(t)-} \left[ \left( \frac{E}{\partial x} - \frac{\partial E}{\partial x} \right) \Psi \right] \, dx - H \Psi \, dt \]

\[ \int_0^{t_f} \int_{\Omega_l} \left( H \frac{\partial \Psi}{\partial t} + E \frac{\partial^2 \Psi}{\partial x^2} \right) \, dx \, dt = \] (C.7)

\[ = - \int_{S(t=0)}^L H(T_{ini}) \Psi(x,0) \, dx + \int_0^{t_f} \frac{\partial \Psi}{\partial x}(L,t) E(L,t) \, dt - \] 

\[ - \int_{S(t)+} \left[ \left( \frac{E}{\partial x} - \frac{\partial E}{\partial x} \right) \Psi \right] \, dx - H \Psi \, dt \]
C.1. Justification

Addition of Eqns (C.6) and (C.7), further subtraction of Eqn (C.5) and considering continuity of $E$, $\Psi$ and $\frac{\partial \Psi}{\partial x}$ across the interface render:

$$\int_{S(t)} \Psi \left( \left[ \frac{\partial E}{\partial x} \right]_s^l \, dt + \left[ H \right]_s^l \, dx \right) = 0 \quad (C.8)$$

Finally, since at the interface $x = S(t)$, it follows that:

$$\left[ H \right]_s^l \frac{dS(t)}{dt} = - \left[ \frac{\partial E}{\partial x} \right]_s^l \quad (C.9)$$

which is the expression of the Stefan condition formulated in extensive variables.
D.1 Theoretical basis

The template-fitting technique was presented first by Kirby and Armbruster (1992) as a pre-processing algorithm to be performed before applying the Proper Orthogonal Decomposition (POD) method in the framework of spatio-temporal complex systems. Later, the method was first included into a dynamical model reduction procedure for advection problems by Glavašky et al. (1998). As a continuation of this work, Rowley and Marsden (2000) extended the technique for generic travelling structures.

The objective of the template-fitting is to remove the variables associated to the symmetry of the problem, this is, the travelling modes. To that purpose, the strategy proposed consists of shifting the system data so that the best match to a preselected template is obtained (Rowley and Marsden, 2000), and then perform POD to the new and shifted system.

Thus, given a PDE system with periodic boundary conditions of the form:

\[
\frac{\partial u(x,t)}{\partial t} = D(u(x,t)), \quad \forall x \in [0, L], t > 0 \tag{D.1}
\]

\[
u(0, t) = u(L, t)
\]

where \(D(u(x,t))\) represents a nonlinear spatial operator, and the solution \(u(x,t)\) is such that exhibits translational symmetry:

\[
u(x,t) = u(x + c(t), t) \tag{D.2}
\]

with \(c(t)\) being the shift operator which accounts for the amount of the spatial
translation considered, the application of the \textit{template-fitting} technique will involve the following steps:

1. **Choosing a template** $u^{\text{temp}}(x)$. This will be the solution of the system D.1 at a preselected time.

2. **Determining the shift operator** $c(t)$. In Rowley and Marsden (2000) this has been done by solving an optimisation problem, although alternative ways could be suitable, too.

3. **Shifting the data.** Once $c(t)$ has been obtained it follows the computation of the shifted ensemble of data:

   \[
   \tilde{u}(x, t) = \tilde{u}(x + c(t), t) \tag{D.3}
   \]

   where $\tilde{u}(x + c(t), t)$ is the solution of the shifted reduced order model:

   \[
   \tilde{u}(x + c(t), t) = \sum_{i=1}^{\text{neig}} a_i(t) \phi_i(x + c(t)) \tag{D.4}
   \]

   Runborg et al. (2002) employs the properties of the Fast Fourier Transform (FFT) to that purpose.

4. **Computing the reconstruction operator** $\dot{c}(t)$ as follows:

   \[
   \dot{c}(t) = \frac{\langle D(\tilde{u}), u^{\text{temp}} \rangle_{x}}{\langle \tilde{u}_x, u^{\text{temp}}_{x} \rangle} \tag{D.5}
   \]

   where the subindex $x$ reads for the first spatial derivative. This operator helps to put back into the dynamical system the "removed" travelling structure (Rowley and Marsden, 2000). Thus, it is required in order to obtain the closure of the shifted system.

5. **Solving the shifted reduced model.** This is the system obtained after introducing Eqn D.3 and Eqn D.4 into the PDE defined by Eqn D.1:

   \[
   \frac{\partial \tilde{u}}{\partial t}(x + c(t), t) + \dot{c} \frac{\partial \tilde{u}}{\partial x}(x + c(t), t) = D(\tilde{u})(x + c(t), t), \quad \forall x \in [0, L] \tag{D.6}
   \]
D.2  Example of application

For the sake of illustration, the template-fitting method has been applied to one of the most simple cases of system with translational symmetry, namely a travelling wave defined by a convection-diffusion PDE of the form:

$$\frac{\partial u}{\partial t}(x, t) = w \frac{\partial u}{\partial x}(x, t) + \frac{\partial^2 u}{\partial x^2}(x, t), \quad \forall x \in [0, L]$$  \hspace{1cm} (D.7)

therefore

$$D(u)(x, t) = w \frac{\partial u}{\partial x}(x, t) + \alpha \frac{\partial^2 u}{\partial x^2}(x, t)$$  \hspace{1cm} (D.8)

Figure D.1 shows the original data and the shifted system obtained from them.

![Figure D.1](image1.png)

Figure D.1: a) Original solution of the system. b) Shifted solution of the system. Please note that the chosen template is the initial data (blue solid line).

![Figure D.2](image2.png)

Figure D.2: Comparison between the full model (solid) and the ROM solution (marks).

In addition, the resulting reduced solution achieved by employing only 2 POD basis
(from the 128 equations solved in the full system) once the shifting has been undone is shown in Figure D.2. As can be seen, there is a very good agreement when compared to the original full model.
E.1 Matlab piece of code for gain computation in Section 7.4.3

% Parameters for the observed system
ndisc = 101; % Number of spatial nodes
neig = 30;  % Total number of modes in the ROM
p = 21;     % Number of spatial measurements
nobs = 10;  % Number of modes to observer

% Initializing the measurement operator
Pm = zeros(p,ndisc);

% equally espaced measurements-- suboptimal location
idx_m = round(linspace(1,ndisc,p));

% Building the measurement operator
for ii = 1:p
    Pm(ii,idx_m(ii)) = 1;
end

% Defining the matrix Q=Phi’P'
Q = Phiobs’*Pm’;
% Computing the gain through Lyapunov equation

\[ QQt = Q*Q' \]

% Matrix stability norm

\[ \text{alpham} = \text{norm}(<\lambda>) \]

% Lipschitz constant

\[ \beta = 100 \]

% Constant

\[ \eta = 0.5 \]

% Minimum eigenvalue of matrix QQ'

\[ \lambda_{\text{d}} = \text{min}(<\text{eig}(QQt)> \]

% Maximum eigenvalue of matrix QQ'

\[ \lambda_{\text{u}} = \text{max}(<\text{eig}(QQt)> \]

% Computation of the Lyapunov matrix P

\[ \alpha = 1.1*(\text{alpham} + \beta*\lambda_{\text{u}}/((1-\eta)*\lambda_{\text{d}})) \]

\[ B = \lambda_{\text{d}} + \alpha*\text{eye}(\text{size}(<\lambda_{\text{d}}>)) \]

\[ P = \text{lyap}(B,-QQt) \]

% Inverse of P

\[ iP = P\text{\textbackslash}eye(\text{size}(P)) \]

% gain definition

\[ \omega = iP*Q \]

% gain tuning

\[ \omega = \omega/5e8 \]
Resumo

Motivación e obxectivos

Tanto na industria alimentaria como na biotecnolóxica a utilización de procesos térmicos está amplamente estendida. Con todo, os procedemento de toma de decisións e o seu propio funcionamento están aínda rexidos por protocolos bastante ríxidos, que son difíciles de adaptar a cambios bruscos nas condicións da producción ou a trastornos no proceso causados por acontecementos imprevistos (falta de materiais, fallos mecánicos, etc).

Ademais, debe considerarse tamén que, na maioría dos casos, as políticas de operación que se seguen nestes procesos non foron deseñadas de acordo con principios óptimos, e polo tanto, existen amplos marxes de mellora non só nos custos de operación (que sería desexable sempre reducir), senón tamén na calidade do produto final.

Neste ámbito o desenvolvemento e implantación de modelos matemáticos orixinados a aplicacións en tempo real constitúe un recurso de grande interese. Estes modelos serían empregados como o núcleo de ferramentas e métodos computacionais desenvolvidos para o control en liña de procesos industriais que garantirían a flexibilidade de operación desexada.

Son moitas son as vantaxes relacionadas coa modelaxe de procesos que se poden enumerar e van dende proporcionar un entorno virtual onde avaliar novos procedemento e equipos, ata prever os efectos de fallos ou mal funcionamento da planta na de calidade do produto. Porén, os requisitos específicos demandados pola implementación de tarefas en tempo real esixen modelos de baixa dimensión, facilmente
manexables desde el punto de vista computacional y que permiten reducir los tiempos de cálculo asociados a su resolución numérica.

En respuesta a esta necesidad surge el concepto de modelo operacional, que debe ser entendido en el sentido de un modelo obtenido a partir de primeros principios, que se simplifica aproveitando tanto las características físicas del sistema como empleando métodos matemáticos de reducción de modelos, y que representa de forma fidedigna el comportamiento del proceso.

Por tanto, a la hora de describir los objetivos de esta tesis de doctorado se tiene en cuenta, por una banda, el desarrollo y detalle de una metodología que permita la flexibilidad requerida en la producción a nivel industrial; además también se pretende mostrar la aplicabilidad y ventajas de este enfoque integral de modelado por medio de ejemplos basados en aplicaciones con real de diferentes procesos de interés para las industrias de alimentos e/ou biotecnológicas.

Introducción

A mayoría de los procesos de interés para las industrias alimentaria y biotecnológica involucran variables de estado (tales como temperaturas y concentraciones) o parámetros que dependen tanto de la variable independiente temporal como espacial. Esta dependencia espacio-temporal confiere a todos estos procesos (desde reactores tubulares hasta problemas de frontera móvil) la súa naturaleza de sistemas distribuidos.

En modelado matemático de sistemas distribuidos se utilizan sistemas acoplados, normalmente no lineales, de ecuaciones en derivadas parciales (EDP), que pueden ser derivados a partir de los denominados primeros principios, esto es, a partir de las leyes de conservación de energía, masa y momento. Estas características de los sistemas distribuidos hacen que sean complicadas, en general imposibles, la obtención de soluciones analíticas, o que sea necesario el empleo de métodos numéricos para a su resolución.

Aunque no es el objetivo de esta tesis ofrecer una descripción detallada de los métodos numéricos empleados en la resolución de sistemas de ecuaciones en derivadas parciales (EDP), en la Parte I esta memoria está dedicada a ofrecer una visión general de estas técnicas numéricas y de sus características fundamentales. Además, en esta primera parte se introduce también las herramientas computacionales empleadas para su implementación numérica, y que serán utilizadas más adelante.

Como se acaba de apuntar, en la Parte I se introducirá a modelado de sistemas distribuidos y técnicas para obtener las suyas representaciones de baja dimensionalidad.
A continuación, esta base será empregada na derivación modelos operacionais para os sistemas que constitúen o obxecto de estudo desta tese. Estes modelos operacionais teñen a función de proporcionar non só representacións precisas dos sistemas, senón tamén computacionalmente eficientes, xa que van ser empregados para aplicacións en tempo real.

A estratexia xeral na obtención dos modelos operacionais para os distintos sistemas presentados comeza co desenvolvemento dunha descripción matemática detallada da dinámica do sistemas baseada en primeiros principios. Unha vez que queden aseguradas as capacidades predictivas desta primeira representación, o seguinte paso será o emprego de técnicas de redución de modelos. Neste marco, dous enfoques distintos, físico e matemático van ser combinados. Deste xeito, e fundamentadas nun coñecemento do sistema, algunhas simplificacións e suposicións deben ser adoptadas co fin de capturar as características suficientes do sistema necesarias para reproducir o seu comportamento con rigor. Ademais, tamén se aplicarán técnicas matemáticas de redución de modelos para rebaixar aínda máis a dimensionalidade do sistema, diminuíndo os tempos computacionais e conservando a precisión desta representación.

Así pois, os métodos numéricos clásicos empregados para resolver ecuacións en derivadas parciais (EDP), tales como o Método dos Elementos Finitos (MEF) ou Método de Diferenzas Finitas (MDF), serán descritos no Capítulo 1. Estes métodos fan uso dunha discretización do dominio espacial para aproximar a correspondente solución do sistema mediante funcións de base locais. Dependendo do tamaño do dominio discreto empregado, e do número de ecuacións que deben ser resoltas, estes métodos poden resultar computacionalmente custosos, e non aseitados para as aplicacións en tempo real.

Como alternativa aos métodos clásicos xorden os Métodos de Orde Reducida (MOR), unha ferramenta moi útil para reducir a dimensión (graos de liberdade) dos sistemas. Mediante a captura de soamente as dinámicas máis lentas do sistema, pode reterse a esencia do comportamento do mesmo, que será reproducida nun espazo de baixa dimensión adecuado para aplicacións en liña. O Capítulo 2 presenta estas técnicas de redución de modelos, introduciendo dous dos seus máis representativos exemplos: a Descomposición Espectral do Laplaciano (LSD) e Descomposición Ortogonal Propia (POD). Do mesmo xeito, introducirase tamén o uso das matrices de Elementos Finitos na súa implementación numérica, todo mediante o emprego dun exemplo práctico de aplicación.

Por último, a modelaxe de problemas de fronteira móbil, que constituén un caso
particular de sistemas distribuídos, presentase no Capítulo 3. Este tipo de sistemas presenta domínios espaciales que varián no tempo, nos que polo menos unha fronteira (que pode ser interior) forma parte do conxunto de incógnitas do problema, e da que deben ser determinados, como unha función do tempo e do espazo, a súa posición e/ou velocidade. Como non hai moitas solucións analíticas dispoñibles, os principais retos que deben afrontarse ó tratar este tipo de problemas están relacionados co desenvolvemento de esquemas numéricos precisos. Mediante a resolución dun exemplo ilustrativo van ser introducidas as características esenciais dos métodos empregados para resolver problemas de fronteira móbil. Tamén se vai analizar a aplicabilidade dos métodos de redución do modelos en domínios variables dependentes do tempo.

Unha vez introducidas as ferramentas a empregar no desenvolvemento do esquema de modelaxe proposto, a Parte II da tese dedicase a presentar, mediante exemplos ilustrativos, as capacidades e vantaxes desta metodoloxía.

No Capítulo 4 preséntase o modelo operacional desenvolvido para describir o funcionamento dun reactor intercambiador de calor no que ten lugar unha reacción altamente exotérmica. Neste capítulo descrebense en detalle os pasos a seguir no proceso de modelaxe, e complementariamente, tamén se presenta o problema de control óptimo para a posta en marcha do reactor, atendendo a restricións de seguridade relacionadas coa temperatura no interior do equipo como exemplo de aplicación en tempo real.

Por outra banda, os Capítulos 5, 6 e 7 están orientados á aplicación do esquema de modelaxe proposto no caso particular anteriormente comentado dos problemas de fronteira móbil. En concreto, van ser abordados aqueles problemas caracterizados pola existencia dunha fronte en movemento asociada a un cambio de fase, é dicir, os denominados problemas de Stefan.

No Capítulo 5 vaise presentar un modelo baseado nunha análise pormenorizada das escalas de tempo para procesos de liofilización, namentres que o Capítulo 6 está dedicado ao control óptimo do proceso e ao desenvolvemento dunha nova metodoloxía para a estimación en liña de forma non invasiva da temperatura da fronte. Finalmente, no Capítulo 7 vaise aplicar a transformación en variables extensivas (entalpía e variable de Kirchhoff) non só ao caso do problema de liofilización, senón tamén a un problema de cambio de fase xenérico a fin de introducir a aplicación de técnicas de redución de modelos no ámbito dos problemas Stefan.
Conclusións

O traballo presentado nesta tese está centrado no desenvolvemento de modelos operacionais axeitados para aplicacións en tempo real de procesos distribuídos, con particular énfase en problemas de fronteira móbil de especial interese nas industrias alimentaria e biotecnolóxica.

O concepto de modelo operacional responde a aqueles modelos matemáticos, baseados en primeiros principios, que se derivan de considerar simplificacións físicas (apoiadas no estudo e comprensión do sistema) e técnicas matemáticas de redución de modelos. Esta metodoloxía de modelaxe achega unha representación de baixa dimensionalidade (ao tempo que precisa) do sistema, e grazas a cal é posible reducir de forma significativa os tempos de cálculo computacional empregados na súa resolución numérica. Esta característica resulta clave cando se abordan problemas de control óptimo ou estimación en liña de parámetros.

Os conceptos xerais relacionados coas técnicas de modelaxe de procesos de sistemas distribuídos aparecen na Parte I desta tese. As técnicas clásicas de resolución de sistemas de ecuacións en derivadas parciais, tales coma o Método de Elementos Finitos (MEF) ou o Método das Diferenzas Finitas (MDF) son descritos nese primeiro bloque introdutorio. Ademais, aí ofrecese tamén unha revisión doutros métodos alternativos aos clásicos que se empregan para a redución dimensional dos modelos aproveitando a propiedade disipativa dos sistemas. Entre estes últimos atopanse a Descomposición Ortogonal Propia (POD nas súas siglas inglesas) e a Descomposición Espectral do Laplaciano (LSD).

Do mesmo xeito, a Parte II está dedicada a ilustrar, mediante de dous casos de traballo, as vantaxes do esquema de modelaxe orientado a aplicacións en tempo real.

O primeiro deles exemplifica o desenvolvemento dun modelo operacional para o caso particular dun reactor intercambiador de calor de tipo compacto. Os inconvenientes xorden en relación á reacción altamente exotérmica considerada e a dificultade que supón manter a temperatura do reactor entre os valores de operación de marcados polas normas de seguridade.

Para este primeiro caso de estudio derivouse o modelo operacional do denominado Open Plate Reactor (OPR). O seu comportamento foi validado por comparación con traballos previos existentes na bibliografía. En xeral, este modelo proposto reproduce adecuadamente o comportamento esperado do sistema. Con todo, atopáronse certas diferenzas con respecto aos resultados xa existentes na literatura revisada, e que foron
pertinentemente comentadas e xustificadas.

A continuación, dúas técnicas diferentes de redución de modelos foron aplicadas a fin de obter unha representación de baixa dimensionalidade para este sistema representativo do Open Plate Reactor (OPR): a Descomposición Espectral do Laplaciano e a Descomposición Ortogonal Propia. Atopouse que o poder de redución da Descomposición Espectral do Laplaciano está fortemente determinado polo valor do número Péclet característico do reactor. Para poñer este feito en evidencia, comparáronse dous sistemas con distinto valor de Péclet: o primero deles correspondente a un valor alto e o segundo representativo dun valor baixo. Os resultados obtidos amosaron que aqueles sistemas nos que o mecanismo de difusión é máis importante, isto é os que teñen un número de Péclet baixo, son os más adecuados para a aplicación da técnica de Descomposición Espectral do Laplaciano.

Ademais, continuando coa avaliación das técnicas de redución de modelos, achouse que o método da Descomposición Ortogonal Propia constitúe unha ferramenta efectiva á hora de obter o desexado modelo de orde reducida para o reactor OPR, pois acadouse unha diminución de ata o 50% no tempo computacional empregado para resolver o modelo en comparación co tempo empregado ó utilizar o Método dos Elementos Finitos (modelo completo). Tamén se puxo de manifiesto que a redución de modelos baseada no emprego da Descomposición Ortogonal Propia e totalmente independente da importancia do fenómeno difusivo (isto é, independente dos valores do Péclet).

A representación de baixa dimensionalidade obtida mediante Descomposición Ortogonal Propia foi empregada como núcleo da estratexia de control óptimo presentada no Capítulo 4. Trazáronse dous escenarios diferentes para tentar acadar o obxectivo seguinte: realizar unha posta en marcha do reactor satisfacendo os requisitos de seguridade ó tempo que se maximizaba a conversión de reactivos. O primeiro caso proposto consistía nun problema de optimización dinámica sen restricións, namentres que o segundo introducía restricións sobre as variables de estado. A utilización do esquema de orde reducida permitiu diminuír considerablemente o tempo computacional (unhas 1000 avaliacións da función obxectivo realizanse en 30 segundos), ó tempo que facilitou a análise do sistema.

Por outra banda, o segundo caso de traballo centrouse na aplicación do esquema de modelaxe proposto a problemas de fronteira móbil, especificamente aqueles que involucran unha fronte en movemento asociada a un cambio de fase: os denominados problemas de Stefan.
Resumo

Este tipo de modelaxe conceptual revelouse como unha ferramenta moi conveniente para acadar un mellor entendemento da dinámica do proceso de liofilización de alimentos e da súa influencia na duración do ciclo e na evolución histórica da temperatura do sistema, que son considerados os parámetros clave que determinan a calidade do produto, a súa estabilidade e a produtividade da operación.

Tamén para este segundo caso se derivou un modelo baseado en primeiros principios e facendo uso dunha rigorosa análise das escalas de tempo presentes no proceso. Dita análise se valeu das propiedades termofísicas inherentes do material co obxectivo de definires as escalas de tempo involucradas na operación de liofilización.

Esta reducción baseada nas escalas de tempo fixo posible ter en conta soamente o fenómeno de interese, isto é a distribución de temperatura no producto ó longo do tempo, e obviar outros mecanismos que ocorrían a diferentes escalas. Obtívose así un modelo de baixa dimensionalidade e orientado ó control en tempo real, que foi validado empregando datos da bibliografía existente.

O modelo na escala da matriz foi empregado como núcleo da estratexia de control óptimo proposta, que definiu as condicións óptimas de operación para minimizar o tempo do proceso liofilización garantindo a calidade do produto (contido final de auga) mediante a resolución dun problema NLP dinámico.

Analizáronse distintos escenarios de control. Primeiramente, consideráronse perfís constantes para as variables de control $T_L$ e $P_c$ ó longo de todo o horizonte de tempo do proceso, observándose violacións nas restricións impostas sobre a temperatura que poderían levar ó colapso da estrutura porosa do produto. Para este escenario, a calidade final do produto asegurouse a expensas dun incremento do 29% no tempo de ciclo en comparación co perfil constante non óptimo de referencia ($T_L=263K$ e $P_c=10Pa$).

A continuación, ensaiáronse perfís de control variables no tempo, que resultaron ser adecuados para reducir o tempo do proceso naméntre que se mantiña a calidade do produto nos níveis requisídos polo mercado. Os resultantes perfís de control óptimos para $(T_L, P_c)$ permitiron reducir un 17.71% (caso con unha variable de control - $T_L$) e un 25.5% (caso con dúas variables de control - $T_L$ e $P_c$) ó tempo dun ciclo de operación en comparación co caso definido por controles constantes ($T_L=251.75K$ e $P_c=25.398Pa$).

Presentouse tamén unha novidosa metodoloxía deseñada para estimar en líña a distribución de temperatura do produto durante o secado primario do proceso de liofilización. Esta metodoloxía proporciona ademais estimacións fiables da temperatura
Resumo

da frente de sublimación e da súa posición mediante un esquema de tres pasos:

- Os denominados Pressure Rise Tests (PRT) empregáronse para obter estimacións de calidade da temperatura da frente.
- A transformación de Landau foi tamén aplicada ó problema orixinal de fronteira móbil para conseguir unha representación equivalente do sistema nun dominio espacial fixo. As estimacións proporcionadas polo paso anterior do esquema (PRT) utilizáronse aquí para pechar o sistema en derivadas parciais formulado no novo espazo de traballo.
- Redución do modelo mediante Descomposición Ortogonal Propia, que permitiu diminuír a dimensionalidade do sistema e reduciu os custos computacionais do sistema transformado.

Leváronse a cabo experimentos de simulación, e os seus resultados foron comparados coas solucións obtidas para o sistema completo obtidos mediante un software comercial de Elementos Finitos (COMSOL Multyphysics©), que emprega un algoritmo ALE na resolución do problema de fronteira móbil. As estimacións obtidas amosáronse en boa concordancia cos resultados obtidos para o sistema completo. Sen embargo, detectáronse ligeiras diferenzas na parte final do proceso ($t > 30h$), debido a que o tempo requirido para acadar o equilibrio vese aumentado a consecuencia da diminución do fluxo de vapor nesta etapa do proceso.

Finalmente, aplicouse o método baseado na transformación en Entalpía e Kirchhoff a dous problemas de Stefan distintos. O primeiro deles consistiu nun problema de cambio de fase xenérico, onde as capacidades desta técnica foron postas de manifesto. Resolvéronse tanto o problema unidimensional como o correspondente problema en dúas dimensións, incluíndo un caso no que a frente do cambio de fase presentaba unha forma irregular.

O segundo problema tratado, que se correspondía cun problema industrial real (a loiofilización dun produto lácteo), serviu para poñer de manifiesto as dificultades asociadas a este método cando a configuración do sistema non permite correspondencias un a un entre as variables intensivas e as variables extensivas. Aínda así, conseguiuse desenvolver un algoritmo para resolver problemas en 1D e 2D para aqueles caso nos que non se considere a curvatura da frente.

Ademais, obtívose a correspondente representación de orde reducida para o primeiro escenario presentado (problema xenérico). A Descomposición Espectral do Laplaciano

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fue la técnica empleada en este caso. A pesar de que los efectos del fenómeno de Gibbs se manifestaron en forma de oscilaciones espúrias, se presentaron diversas estrategias encaminadas a minimizar su influencia en los resultados obtenidos.

La primera de estas estrategias se basó en una aproximación smooth de la variable discontinua en combinación con una técnica de filtrado. Se obtuvo una reducción de alrededor del 50% en el número de grados de libertad del sistema, estando el modelo reducido en buena concordancia con el comportamiento del modelo completo. Sin embargo, se observó que la exactitud de estos resultados en comparación con el modelo real (sin smoothing) dependía del intervalo $\Delta T$ empleado para aproximar el salto de energía en la variable entalpía.

Para resolver esta dificultad, se desarrolló un observador basado en el modelo reducido del sistema que no requería la aproximación smoothing de las variables intensivas. Se utilizó un esquema de estimación en línea, en combinación con el modelo de orden reducido obtenido mediante LSD, para la reconstrucción de la variable entalpía mediante un número limitado de medidas experimentales. A pesar del fenómeno de Gibbs, los resultados obtenidos mostraron una buena correspondencia con el sistema real, y en el tiempo que se acercó una importante reducción del sistema (aproximadamente del 60%).

Finalmente, se presentó un sencillo ejemplo que debe ser tomado como paso previo en la aplicación de la técnica de template-fitting para la formulación en variables intensivas de los problemas de Stefan.
Contributions

Articles in SCI journals


Articles in preparation

• Model reduction of Stefan problems: an approach in extensive variables. **Lopez-Quiroga, E.** Vilas, C., Alonso, A.A. Process Engineering Group, IIM-CSIC.


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