1 Introduction

Porcelain tiles are a type of ceramic tile that is composed of clays, feldspars, and several other inorganic raw elements, as per its established specification. The primary objective of these materials is to offer comprehensive coverage for both floors and walls. They may be produced using several techniques, such as pressing, extrusion, or other analogous manufacturing processes. The production process encompasses a significant level of grinding, a substantial proportion of raw material melting, and a considerable degree of densification following the burning process. These procedures yield products characterized by minimal porosity and exceptional technical capabilities. The tiles have the potential to be coated with enamel, remain uncoated, undergo polishing, or retain their natural finish, and may or may not be repaired.

Due to their high density, complete vitrification, and low porosity, porcelain tiles are considered a component of traditional ceramics. The ultimate overall porosity of the product varies between 3% and 6%, with less than 1.2% representing open porosity specifically linked to water uptake. Based on the ISO 13006:2018 standard (International Organization for Standardization ISO standard, 2018), it is recommended that the water uptake rate not exceed 0.5%.

The highest added value among traditional ceramic products is found in porcelain tiles, mainly due to their technical and aesthetic characteristics. The highlighted properties of the product include high mechanical strength, high wear resistance, and a high gloss when polished. A specific microstructure is required to achieve the aforementioned characteristics. A vitreous phase that disperses both newly formed phases (mullite) and residual minerals (quartz and feldspars) is what distinguishes porcelain tiles. A glassy phase is usually 40%-75 wt.% of the matrix, with quartz particles 10-30 wt.%, mullite crystals 4-10 wt.%, and unmolten feldspar 0-15 wt.% dispersed on it (De Noni Jr. et al., 2022). This quantitatively dominant vitreous phase governs viscous flow sintering and has a significant impact on the geometrical, mechanical, tribological, and functional properties of tiles (Zanelli et al., 2011). According to De Noni Jr. et al., 14% to 17 wt.% mullite is best suited for achieving a robust microstructure in porcelain tiles manufactured using standard industrial-scale technology (De Noni Jr. et al., 2022). To achieve this characteristic, it is necessary to prepare a mixture of compounds. A triaxial mixture of clay or kaolin, quartz, and feldspar is used to create the starting composition. Fig. 1-1 illustrates the triaxial diagram composition with different ceramic products, including porcelain tiles:

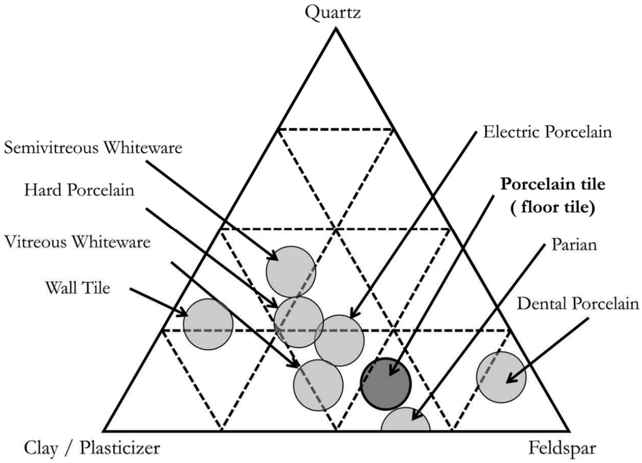

Fig. 1-1: Diagram of triaxial composition of ceramic products compared to the composition of porcelain tiles.

The clay fraction contributes plasticity and dry mechanical strength during processing, and it develops mullite and glassy phases during firing. Feldspars (primarily sodium feldspar) form glassy phases at low temperatures, aiding the sintering process and allowing for almost no (0.5%) open porosity and a low level of closed porosity (10%). Quartz promotes thermal and dimensional stability due to its high melting point (Sánchez et al., 2010). Both quartz and feldspars are materials that do not provide plasticity in the presence of moisture; they correspond to the fraction of larger particles in the mixture, normally with maximum sizes of up to 63 µm (De Noni Jr. et al., 2009). This is why they favor packing the particles during the compaction phase and water evaporation during the drying of the components. Furthermore, the crystalline particles produced by these substances or crystallized during firing significantly contribute to the strength of the microstructure. Additionally, additives such as talc, calcite, dolomite, and diopside are examples of strong fluxing additives to reduce the sintering temperature (Dondi, 2018).

The global porcelain tiles market is rising rapidly. It is projected to grow from $54.41 billion in 2021 to $77.82 billion in 2028 at a compound annual growth rate of 5.2% (Market research report, 2022). In 2017, global ceramic tile production was about 13500 million m^2 (Ferrer et al., 2019). Currently, Brazil is the third largest producer and consumer and is alone responsible for an increase in production of 4200% from 4 million m^2 in 2001 to 168 million m^2 in 2020 (Bombazaro and Bernardin, 2022). In Europe, Spain is currently (2002) the leading manufacturer, increasing tile production by 20.3% (to 567 million m^2) when compared to 2021.

1.1 General description of manufacturing sequence

A series of processing steps are necessary to produce porcelain tiles. There are two main alternative process configurations, so-called “wet” and “dry” routes (De Noni Jr. et al., 2010). Fig. 1-2 depicts the “wet” manufacturing route for porcelain tiles.

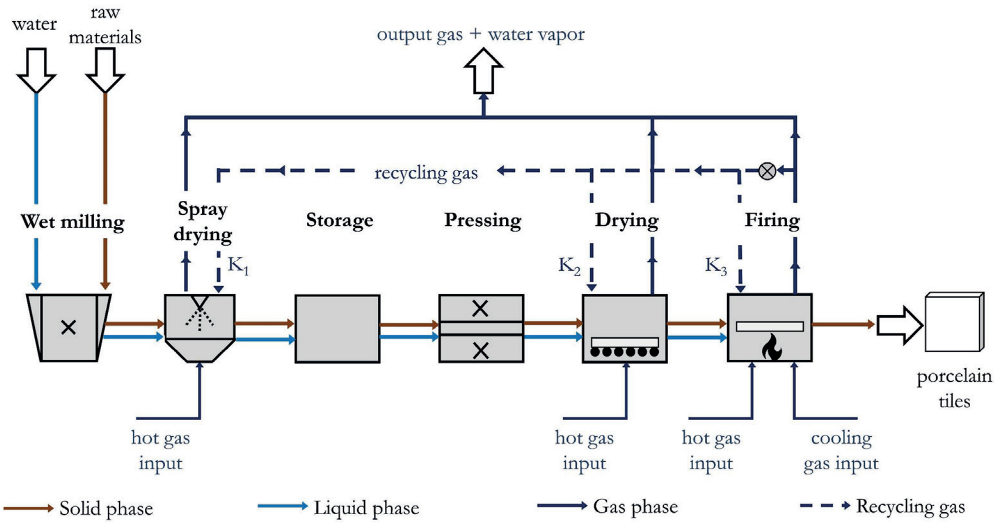

Fig. 1-2: Flowchart of porcelain stoneware showing the main manufacturing steps of the "wet" route. Image previously published in (Alves et al., 2023a).

Three main stages can be distinguished: (1) preparation of the powder, from the grinding and homogenization of the raw materials by wet milling, with subsequent spray drying of the resulting suspension; (2) shaping, usually by pressing from atomized powder, with moisture content between 5 and 7% and pressure of compression between 35 and 50 MPa; (3) firing, in a rapid cycle between 40-60 min cold-to-cold and maximum temperature between 1180 and 1220 ºC, determined by obtaining the maximum densification and content of water uptake <0.5% or <0.1%.

The "wet" route is basically mandatory when the raw materials have very different physical characteristics and must be homogeneously mixed in addition to being grinded. This is the case for the production of porcelain tiles. However, because of the excellent granulometric characteristics of the powder obtained, the masses prepared by the wet process and thus atomized allow for better filling of the matrices or dies during the pressing stage. This results in higher green and dry product densities and therefore better board mechanical properties, both before and after firing. These advantages are very important, above all in single-firing technology, in particular when rapid firing cycles are used (Oliveira and Hotza, 2015). The main benefit of the dry process is that it is significantly less expensive to run from an energy standpoint (Agrafiotis and Tsoutsos, 2001).

During wet milling, the initial raw materials are mixed with water and grinded to reduce their particle size. The process is typically carried out in a rotary drum ball mill using high-grade alumina balls that come in different sizes to improve gridding efficiency (Breitung-Faes and Kwade, 2013). The gridding elements total 50% of total volume of the mill, and the velocity ranges from 50-65% of the total critical speed (Reed, 1995). The critical speed, which is mathematically determined, is the speed above which the centrifugal force prevents the cascading movement of the grinding elements, which reduces the effectiveness of the grinding process (Oliveira and Hotza, 2015). The grinding process can last from 3 to 8 hours, depending on the type of formulation. For porcelain tile, this time often exceeds 10 hours.

The drying of the obtained slurry to obtain a powder with the desired granulometry and humidity characteristics is done using spray-dryers, also known as atomizers. A controlled evaporation process removes the water during atomization. The water removal is not complete, and some residual water must remain in the formed granules to provide adequate plasticity for the compaction stage (Oliveira and Hotza, 2015). The remaining moisture on the granules varies according to the characteristics of the raw materials, ranging from 4 to 8 wt.%. High-pressure pumps (25 to 30 bar) are used to inject the slurry, usually from the bottom up (Ribeiro et al., 2001). The nozzles to inject the slurry are normally made of metal and distributed in the ring in various diameters (2 to 4 mm), depending on the desired particle size distribution. The generated steam from auxiliary gas burners is injected at the top of the atomizer and distributed tangentially at temperatures ranging from 650º to 750ºC , depending on the desired productivity and final characteristics of the atomized powder (Negre et al., 2000; Oliveira and Hotza, 2015). The heat exchange normally occurs in the opposite direction from the injection of the slurry. As a result of the drying, granules (agglomerates) with varying particle sizes and a certain percentage of moisture are formed. The morphology of the granules can be either spherical or with internal holes, giving rise to the term "donnut" morphology.

The "donnut" morphology formation is fundamentally due to four mechanisms (De Noni Jr., 2005; dos Santos Conserva et al., 2017):

- The formation of an elastic film of low permeability around the pulverized particles, which reduces the evaporation speed and gives rise to an increase in temperature inside the atomized particles, causing their "swelling";
- Presence of soluble salts, which precipitate on the surface of the grains after migration to the periphery of the particles and evaporation of the water;
- Migration of the liquid containing insoluble solids (typical case of clay suspensions) to the surface of the atomized grains under the action of capillarity, dragging the solid particles. The subsequent evaporation creates an internal porosity in the granules;
- Retained air in the slurry.

After spray drying, the obtained granules are transported through conveyor belts to the storage silos, where they remain for at least 48 hours to homogenize, cool down, and stabilize the moisture content (Amorós et al., 2002). The utilization of granules with temperatures exceeding 40˚C results in various complications during the processing phase. Elevated temperature negatively impacts the flowability of the granules within the die, impeding the proper filling of the die cavity. As a result, this can cause loading failures and errors in the geometric characteristics of the final tile. Furthermore, elevated temperatures can cause cracks to form prior to reaching the firing unit, thereby impacting the geometric stability of the end product. Gradients in temperature and concentration are what drive the moisture change (loss or gain) and redistribution during this stage. Those effects and the porous intricate structure are three main aspects to consider in this coupled heat and mass transfer phenomena augmenting the system complexity. Variations in ambient conditions lead to moisture movement. Water evaporates from hot areas, moves across the gas-filled pores by diffusion, and condenses in the colder regions, thus releasing its latent heat of vaporization.

Following compaction, the granules turn into green tiles. Due to its productivity, uniaxial pressing is the most widely used forming method in the production of porcelain tiles. It consists of applying pressure in only one axial direction through a rigid punch to compact the atomized powder contained in a rigid cavity (Albaro, 2000b). The movable base, also referred to as the bottom punch, and the walls, known as the mold die, make up the rigid cavity. Pressure is applied through the upper punch, which is introduced into the cavity containing the powder, formed by the matrix, and the lower punch, which remains immobile at this stage (Albaro, 2000a). Once the tile is compacted, the upper punch is removed, and the sliding of the lower punch allows the extraction of the part from the mold.

To be properly processed, the granules must have the appropriate properties. These characteristics include: good fluidity and packing, so that during the mold filling stage, the mass quickly falls by gravity and fills all the necessary spaces in a homogeneous manner; humidity and plasticity to allow plastic deformation during pressure application; and mechanical resistance, primarily during part extraction and transport to the next stage of the process (Albaro, 2001; De Noni Jr., 2005). During compaction, four main stages can be depicted (van der Zwan and Siskens, 1982; Dondi, 2021):

1. On the first stage, there is sliding and rearrangement without deformation or breaking of granules. A large volume reduction of the material occurs, and normally, after this stage, the upper punch, responsible for applying the force, moves back slightly before continuing to the final stage.
2. The second stage is characterized by the fragmentation and plastic deformation of the granules. During this stage, there is a reduction in the volume and size of the intergranular spaces.
3. In order to achieve a denser packing, the sliding and rearrangement of the primary particles during the third stage reduces the volume and size of the intragranular pores.
4. The fourth stage occurs through fragmentation and plastic deformation of the primary particles.

Fig. 1-3*:* depicts a general scheme for the compaction stages of porcelain tile manufacturing:

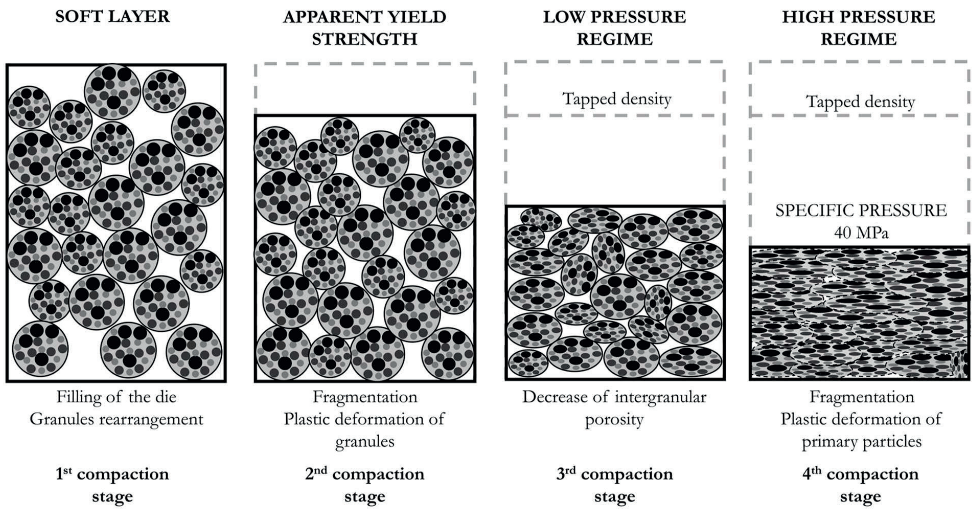

Fig. 1-3: Scheme of the four stages of compacting dried powders. Image previously published in: (Alves et al., 2023b).

The drying process takes place subsequently to the pressing stage, where there is an elimination of the residual moisture of the products from up to 8 wt.% to approximately 1 wt.% (Utlu et al., 2011). Industrially, the drying operation is performed by circulating hot air. The dryers can be either horizontal or vertical. The most common type is the vertical dryer, that consists of a mobile structure built into steel profiles and covered with insulating panels. Mechanically driven tilting systems inside the dryers move the green tiles through the drying channel. Internal pipes distribute hot air with temperatures ranging from 150 to 180ºC to the drying zone and cool air to the cooling zone. Heating of the air occurs through burners that can operate with liquid and/or gaseous fuels. The drying cycles vary from 35 to 70 min (Utlu et al., 2011; Khalili et al., 2014; Oliveira and Hotza, 2015).

The final processing step is firing, where sintering takes place. The purpose of this stage is to transform the green tile into a resistant product. The particles in mechanical contact create continuity of matter in the region of contact, i.e., the particles bond to each other at the atomic level by forming contacts that grow as a function of the transport of atoms or ions to this region (Reed, 1995). The formation of liquid phases and viscous flow in porcelain tiles are the main causes of this transport, which can happen through a number of different mechanisms. In addition to sintering, a series of chemical transformations take place.

Among the physical transformations can be cited (Oliveira and Hotza, 2015):

- Dehydration: loss of moisture from capillarity and adsorbed water;
- Thermal dilatation: variation in the dimensions of the tiles with heating and cooling;
- Allotropic transformations: change in the crystalline arrangement of certain materials. The best known is the transformation of the quartz from α to β at 573ºC.
- Fusion: change of physical state from solid to liquid.
- Sintering: transporting material still in the solid state from any region to the interface region between two or more particles.

Whereas the chemical transformations are:

- Loss of constitution water: water that is part of the crystalline structure of the clays
- Decomposition of organic matter
- Decomposition of carbonates ($CaCO_3 \rightarrow CaO + CO_2$)
- Increase in oxidation state of iron and sulphur oxides
- Formation of new phases as mullite, for example.

The firing process does not simply involve bringing the tiles to a certain temperature. The cycle involving the speed of heating, the residence time at maximum temperature, and the speed of cooling are equally important. Several parameters must be controlled during the stage, such as firing intervals, maximum temperature residence time, temperature uniformity in the kiln, atmosphere of the furnace, consistency in charging the furnace, and dimensional variation of the fired tiles (De Noni Jr., 2005; Oliveira and Hotza, 2015). The cycles and firing temperatures adopted, in general, vary, respectively, between 40 and 60 min and 1180 and 1220 ºC. The optimum sintering temperature is the temperature at which the material transport rate is high enough so that the desired microstructure for a given application is achieved in a relatively short time for industrial practice (Rahaman, 2017). The furnaces, or kilns, are specially equipped for the process. The burners and their combustion chambers are normally sized to operate at high flow rates for combustion products and with low specific heat output. The equipment that is used the most frequently during the firing process is the continuous roller kiln. It consists of a long structure, only the center of which is heated directly. The temperature of the green tiles gradually rises as they approach the hottest area in the center of the kiln as they enter and move slowly through it (Refaey et al., 2015b; Refaey et al., 2022) . When the tiles are finished with the treatment, their temperature drops to almost room temperature. The most energy-efficient roller kiln for the firing process is one that is continuously run because it allows the heat from the cooling phase to be used for all earlier drying processes, further reducing the need for fuel.

The entire processing sequence consumes a large amount of energy. Thermal energy consumes the most energy, accounting for 92% of total energy consumption. (Mezquita et al., 2014a). Spray drying of the slurry accounts for 36% of total thermal consumption, drying of green tiles accounts for 9%, and tile firing accounts for 55% of total energy demand—the highest within the manufacturing process. (Mezquita et al., 2014b). All stages of the processing sequence use electrical energy. Milling consumes the most electrical energy, accounting for nearly 22% of total consumption (Monfort et al., 2010).

The thermal energy required in the process is transferred via hot air, which is typically obtained through natural gas combustion, resulting in carbon dioxide, sulfur dioxide, and nitrogen oxide emissions (Ye et al., 2018), which have an impact on the environment. For example, the amount of CO_2 generated while producing one ton of fired ceramic tiles is estimated at 265 kg (Mezquita et al., 2009). Special attention has been given to the environmental aspects of porcelain tile manufacturing in Europe due to the new 2030 climate and target set by the European Commission and the goal of an 80-95% reduction in CO_2 emissions by 2050 (European Commission; European Commission, 2011). Fuel consumption must decline in order to reduce pollutant gas emissions. This is a highly complex optimization challenge since the factors that improve one step can often harm another stage. Therefore, it is not trivial to achieve environmentally friendly tile manufacturing while minimizing costs, increasing productivity, and maintaining industry competitiveness. Increasing automation can help with process control, allowing for an increase in production volume, ensuring high-quality indicators, locating or removing bottlenecks, and reducing production costs.

Within the context of Industry 4.0, there is a current shift towards completely digitizing manufacturing processes (Erol et al., 2016). The industrial revolution has a significant effect on small and medium-sized enterprises that rely heavily on manual labor (De Noni Jr. et al., 2010; Pitarch Monferrer, 2017). This is the case of the ceramic industry. While ceramic manufacturing processes are extensively automated in terms of product handling and material processing at the level of individual operations, there is a lack of integration among them. The absence of integration presents a constraint for digitized production. This phenomenon is particularly prevalent in the traditional ceramic industry, specifically in the production of porcelain tiles (Pitarch Monferrer, 2017).

There are multiple studies available for each individual process unit of the manufacturing process (Breitung-Faes and Kwade, 2013; Breitung-Faes and Kwade, 2014; Demarch et al., 2014; Kriaa et al., 2014; Gültekin et al., 2017; Soldati et al., 2018a; Ferrer et al., 2019). Additionally, there are experimental studies that investigate the impact of different unit parameters on the final product (Alves et al., 2010; Oliveira, A. P. N. D. and Hotza, 2015; Martin et al., 2016; Soldati et al., 2018b). These studies enable us to achieve a high degree of standardization in these processing steps. Barata et al. (Barata et al., 2019) have identified that the ceramic industry's high level of standardization hinders digitalization, primarily because of the extensive use of experimental material characterization and trial-and-error methods when modifying process parameters. This empirically derived methodology is laborious, costly, and does not facilitate the identification of optimal processing parameters regarding energy or raw material usage, as well as the enhancement of the final product's characteristics. However, due to the complexities involved in simulating the entire process chain, there is currently no existing research on the interdependencies between the parameters of individual units and their impact on other units and overall outcomes within the manufacturing chain. To enhance overall efficiency, it is imperative to consider the complete manufacturing process, which comprises various interconnected steps involving energy and material streams. In a study examining the scientific and technological progress of the porcelain tile industry, Sánchez et al. (Sánchez et al., 2010) identified a constraint in the sector's use of simulation and modeling tools. This limitation persists despite significant technological advancements made by machinery builders over the past three decades. These tools are essential for enabling the practicality of developed processes, the ability to predict outcomes, and increased efficiency through innovation in manufacturing, control, and measurement processes (Pitarch Monferrer, 2017).

Numerical simulations, which have developed into a practical tool to optimize, design industrial equipment, and assess operation parameters, play one of the key roles in Industry 4.0. Flowsheet simulations can be effectively used for numerical investigations of complex processes with the connection of production steps by material and energy streams (Dosta et al., 2020). The flowsheet simulation concept involves solving material and energy balances numerically as well as determining intensive state variables for various process structures (Skorych et al., 2017). Flowsheet simulation tools have become widely accepted in the field of chemical engineering for processes involving fluids. Nevertheless, the application of such instruments remains infrequent for procedures that encompass solid materials, particularly ceramic powders. The development and application of tools for the solid process have only been initiated in recent years (Dosta et al., 2020). One of the reasons is the increased complexity resulting from a detailed description of solids using multi-dimensional distributed parameters. Particles can be distributed along with various property coordinates, such as size, density, moisture content, etc. Therefore, in order to accurately quantify material flows that are described by multidimensional parameters, it is necessary to utilize specialized techniques such as transformation matrices (Skorych et al., 2019a). The simulation framework Dyssol has been proposed for performing dynamic and steady-state flowsheet calculations of solids processes (Skorych et al., 2017; Skorych et al., 2020). This is a versatile system that is compatible with multiple operating systems and can be enhanced with additional models for various unit operations. In this case, model-predictive calculations can be used effectively in flowsheet simulations to plan and improve processes, evaluate operational parameters, and predict more environmentally friendly production of complex plants like those that make porcelain tiles, where many processing units are connected to each other.

1.2 Motivation

The aim of this study is to accurately represent and simulate each individual stage of the porcelain tile manufacturing process sequence, as well as the transfer of data from one step to the next. This encompasses the initial combination phase of the raw material powder and extends all the way to the ultimate manufacturing of the finished product. The aim of this study is to develop a simulation methodology that can be used to optimize the process manufacturing chain and be used as a decision-making tool. In order to achieve this objective, the subsequent intermediate targets were addressed:

- Implementation and validation of macroscale models for the whole process sequence using flowsheet simulation, which connects each individual component of the process;
- implementation of the thermal and electrical energy consumption for the entire process sequence, which includes investigating how various processing parameters affect the energy consumption;
- use the Discrete Element Method (DEM) and the Bonded Particle Model (BPM) to run microscale simulations that will help you understand how different stages of processing work on a micromechanical level. This will aid in the validation and improvement of the semi-empirical correlations employed in macroscale models;
- the development of an optimization methodology to analyse different production scenarios and propose modifications in the manufacturing process aims to reduce energy costs and CO_2 emissions while maintaining product quality.

1.3 Outline

The theoretical basis for the numerical analyses, which encompass flowsheet simulations and DEM-BPM simulations, is presented in Chapter 2.

Chapter 3 is dedicated to model development and numerical background. The semi-empirical models utilized to develop the flowsheet simulations are described in detail in Chapter 3.1. The energy consumption modelling is described in Chapter 3.2. The numerical background of DEM-BPM simulations is elaborated upon in Chapter 3.3 and 3.4, specifically regarding the compaction stage and heat transfer during storage, respectively.

Chapter 4 discusses the materials and methods used in the study. This includes the simulation of large-scale flowsheets (Chapter 4.1), the experiments used to check the DEM-BPM compaction simulations (Chapter 4.2), and the experiments on particle heat transfer analysis used to check the DEM-BPM simulations. The different optimization methodologies and case studies are described in Chapter 5.

Chapter 6 presents the validation of the flowsheet simulation, which includes investigations into sensitivity analysis. The energy consumption validation is presented in Chapter 7 for both electrical energy consumption (Chapter 7.1) and thermal energy consumption (Chapter 7.2). The results and discussion of the different optimization cases are presented in Chapter 8.

The simulation results for the compaction stage using DEM-BPM are presented in Chapter 9. Chapter 10 provides a comprehensive examination of the heat transfer analysis of the storage stage, encompassing both experimental and numerical investigations.

2 Fundamentals of numerical simulations

2.1 Flowsheet simulations

The modeling of integrated production processes requires the consideration of information beyond that obtained from individual process units alone. The behavior of each individual unit can exert a substantial influence not only on subsequent processes but also on the overall process, particularly in the context of recycle streams and the implementation of process control and plant-wide optimization strategies. Consequently, it is imperative to conduct a comprehensive analysis of the integrated system as a whole. The application of flowsheet simulation can be effectively employed for this particular objective.

The process of flowsheet simulation can be conducted in either steady-state or dynamic mode. From a computational perspective, the steady-state analysis offers a simpler approach for simulation and model creation (Dosta et al., 2020). In the field of solids process engineering, it is important to note that not all unit operations demonstrate transient behavior that has a substantial impact on the overall dynamics of the process. This is the case for unit operations with small holdup mass as mills and can be treated as steady-state models (Cotabarren et al., 2013). This is the main reason for employing the steady-state mode in the analysis of this work.

The utilization of flowsheet calculations encompasses four primary applications, namely the modeling of process behavior, sensitivity analysis, process optimization, and process control. One of the key advantages of flowsheet simulations is the ability to model the variation of process parameters throughout the process sequence with the goal of process optimization. Utilizing flowsheet-based sensitivity analysis provides a deeper understanding of how process parameters impact the processing sequence, resulting in the creation of a tool for decision-making (Dosta et al., 2020).

The simulations typically employ empirical or semi-empirical models on which macroscopic states such as mass flow, particle size distributions, and temperatures can be predicted (Dosta et al., 2010; Skorych et al., 2017; Skorych et al., 2019b). However, certain product parameters that have an impact on the overall material characterization are being overlooked. In order to offer a comprehensive depiction of particle systems, it is necessary to employ multidimensionally distributed parameters. As a result, the implementation of a robust process necessitates the utilization of intricate models tailored to specific process units. Transformation matrices have been shown to be useful for accurately changing parameters that are spread out over many dimensions in both steady-state and dynamic calculations (Skorych et al., 2017).

The formulation of transformation laws is specific to each model, resulting in the generation of a transformation matrix instead of the explicit calculation of all output variables. The transformation of