1 Introduction

Many materials that surround us have a particulate form or are produced using processes that involve such materials. According to some studies (Merkus, 2009), about three-quarters of all industrially produced goods have a particular form, including particles of solid (granules, powders, etc.), liquid (emulsions, sprays, etc.), and gaseous (bubbles, foams, etc.) forms. According to others (Schulze, 2014), 60% of all products manufactured by the European chemical industry are estimated to be bulk materials, and a further 20% of products involve bulk materials in production. Other industries that heavily involve or produce granular materials include pharmaceuticals, metallurgy, mining, food processing, agriculture, and ceramics, to name a few.

In many of these industries, to obtain bulk products with desired properties, it is often necessary to combine several stages of material processing, and each stage can be performed on different types of equipment. These steps include, for example, agglomeration, classification, crystallisation, granulation, grinding, or sintering. Examples of such processes include clinker and cement manufacturing (Van Oss and Padovani, 2002; Rahman et al., 2013), crushing plants (Bengtsson et al., 2009; Asbjörnsson et al., 2022), granulation processes (Cotabarren et al., 2015; Diez et al., 2018), tablet manufacturing (Singh et al., 2013; Bano et al., 2022), carbon capture in combustion reactors (Haus et al., 2017), and soil washing (Toebermann et al., 1999).

As a result of advances in these areas, the technological structures of production processes become increasingly complex, leading to corresponding challenges in their design, integration, maintenance, and optimisation. Therefore, it is unsurprising that interest in developing approaches and methods for studying such complex processes regarding their stability, predictability, and dynamic behaviour is only growing. Numerical modelling can make a significant contribution to solving all these problems. However, the need to consider the complex structure of the processes under study makes their modelling a challenging task. The most straightforward approach is to study the different unit operations and sub-steps of the process individually. However, this does not allow us to see the whole picture since, due to the interdependence of individual operations, the work of one block often affects the entire process. As a result, it is necessary to study the entire process as a whole. Flowsheet simulation proved to be a powerful tool for this.

Due to its heterogeneity, the granular nature of the solid phase introduces unique challenges to both unit operation models and flowsheet simulation frameworks. Another critical level of complexity arises from the fact that many solids processing plants include unit operations that exhibit transient behaviour.

Consequently, there is an urgent need for simulation tools capable of capturing the dynamics of these systems. However, despite the importance and diversity of the solids processing, the tools for their dynamic flowsheet simulation remain underrepresented.

This work addresses this gap by focusing on the development of a generally applicable system for the dynamic modelling of solid-phase processes. The proposed framework aims to integrate advanced modelling techniques, which allow the correct treatment of distributed parameters of granular materials and the efficient description of their transient behaviour, into a robust, holistic, and flexible simulation environment.

1.1 Flowsheet simulation

Modern manufacturing processes strive to operate continuously (Dosta et al., 2020), where raw materials are consistently introduced into the system while final products are simultaneously extracted at a similar flow rate, ensuring a steady output of the final goods (Lee et al., 2015). This gives a lot of advantages, such as greater flexibility in the quantity of product produced, better scalability, higher throughputs, optimisation of used space, raw materials, and equipment, better suitability for automation, and even the reduction of waste (Plumb, 2005; Boukouvala et al., 2012). The downside of continuous operation is that due to the rising need to combine different production steps into one composite manufacturing chain, processes become more complex, making them difficult to design, analyse, change, control and optimise. Overcoming this challenge requires better process understanding, which is what flowsheet simulation tools can provide.

Flowsheet modelling, also referred to as process system modelling, is generally understood as a computer-aided technique for simulating the interconnected operations of entire chemical and process engineering plants. It typically involves representing the sequence of the process units, such as mixers, crushers, separators, and reactors, by their mathematical models and connecting them with streams that carry materials between units in the process chain. By solving these models and the corresponding equations of mass and energy balances, engineers can better understand how various individual operations interact and how they affect the overall manufacturing process and the properties of products.

Flowsheet modelling is typically classified as macroscopic modelling. This approach focuses on studying processes as a whole without delving into fine details. For example, macroscopic process modelling does not predict the motion of individual particles or the behaviour of each component within a machine. Nor can it account for the impact of subtle geometric features of the equipment on

the properties of the final product. Instead, it provides the broad, system-wide perspective needed to understand overall process behaviour and performance.

Flowsheet simulations serve multiple purposes across research and development and are also applied to address specific problems within industrial settings. Among the prominent use cases can be distinguished:

- Process planning and design by facilitating initial sizing, estimating equipment costs, and helping in early-stage decision-making (Coker, 2007);
- Process optimisation, aiming to minimise energy consumption, waste production, or raw materials use, to reduce production time, or improve product quality (Wang et al., 2017; Asprion and Bortz, 2018);
- Support in the scale-up of production from laboratory to industrial level by simplifying process validation and regulatory compliance checks (Rakicka-Pustułka et al., 2020);
- Testing hypotheses or investigating the design alternatives, allowing engineers to evaluate “what if” scenarios and assisting them in understanding system responses under various operating conditions (Morais et al., 2010);
- Process analysis in order to gain a better understanding of process behaviour, identify bottlenecks, and assess the sensitivity of the process to specific influences (Schwier et al., 2010; Asprion et al., 2022);
- Support decision-making during the process operation by offering a systematic and quantitative analysis of the process’s responses or by applying strategies of model-predictive control (Cotabarren et al., 2015; Luyben, 2015; Neugebauer et al., 2019).;
- Operator training using simulated scenarios, allowing for the reduction of the risk of equipment damage and enhancing operational safety (Toro et al., 2013; Patle et al., 2014);

Thus, one of the main advantages of flowsheet modelling is its ability to explore complex interactions in interconnected systems without needing physical experiments, significantly reducing costs and time spent. However, it has its limitations. For example, developing models of the unit operations is often a complex task, and even using the modelling tools can be quite challenging (Bezzo et al., 2004). Both tasks are further complicated in the context of dynamic modelling. Accurate flowsheet modelling may require extensive data and accurate parameter estimation, which can be challenging, especially for highly dynamic processes. In addition, high-fidelity modelling can be computationally demanding, and simplifications made to reduce model complexity or improve simulation performance can lead to discrepancies between modelling results and actual data.

Given their advantages and broad applicability, flowsheet simulations are utilised across various industries and for numerous processes: food processing (Ureta and Salvadori, 2023), mixing of pharmaceutical powders (Dias et al., 2023), tablet manufacturing (Boukouvala et al., 2012; Gavi and Reynolds, 2014; Nagy et al., 2021); dry granulation (Park et al., 2018), wet granulation (Metta et al., 2019), biopharmaceutical production (Malinov et al., 2024), porcelain manufacturing (Alves et al., 2021), battery recycling (Punt et al., 2023), carbon capture (Haus et al., 2018), petroleum refining (Azad et al., 2016), biomass gasification (Inayat et al., 2020), to name a few.

1.2 Complexity of solids

As solid particle processes, one defines those that involve solids unit operations, such as crystallisation, drying, granulation, filtration, milling, or sieving within the process sequence. This definition applies regardless of whether the solids are feed materials, intermediates, or final products (Rajagopal et al., 1992). Many studies emphasise the need to distinguish between the modelling of processes involving fluid and solid materials since the latter are more complex in nature, more complicated to handle, and require different, often more sophisticated algorithms and processing methods (Rajagopal et al., 1992; Toebermann et al., 1999, 2000; Werther et al., 2004; Hartge et al., 2006; Pogodda, 2007; Dosta et al., 2010, 2020; Dosta, 2013; Boukouvala et al., 2012).

In flowsheet modelling, liquids and gases are generally described using fundamental principles of thermodynamics and fluid dynamics, applying straightforward equations to calculate properties like density, temperature, and pressure. In contrast, modelling solids demands consideration of additional influences, including particle size, particle shape and roughness, bulk compaction, segregation, breakage, attrition, etc. (Rhodes and Seville, 2024). Subtle differences in these properties and slight variations in process conditions or equipment performance can lead to considerable differences in process behaviour (Muzzio et al., 2002) due to effects of segregation (Umbanhowar et al., 2019), clogging (Cervantes-Álvarez et al., 2023), jamming (Behringer and Chakraborty, 2019), undesired breakage (Reynolds et al., 2005) and others. All this makes bulk materials susceptible to higher degrees of variability, complicating their modelling due to the need to introduce a significant number of new parameters and due to the emerging nonlinearities and discontinuities, which in turn complicate the convergence and accuracy of calculations.

Considering characteristic values of bulk properties like size, abrasiveness, hardness, or moisture content is often insufficient (Toebermann et al., 2000; Asbjörnsson et al., 2022), leading to the introduction of distributed parameters to describe these properties. In most cases, the particle size distribution is the primary and

most important distributed parameter of the solid phase (Hill and Ng, 2002). However, additional distributions can also be critical to a particular piece of equipment. For example, porosity and saturation are essential in agglomerators (Iveson et al., 2002), while particle shape and orientation are critical in crystallisers (Kovačević et al., 2017). Similarly, yield strength is significant in granulators (Liu et al., 2000), moisture content is important in dryers (Alaathar et al., 2013), and chemical composition is essential in various pharmaceutical processes (Metta et al., 2019). Therefore, a solid-state processing equipment model must be able to consider these interconnected distributed parameters, making its development and calculation quite challenging.

Even describing such distributed parameters presents a unique challenge. Granular materials typically consist of individual heterogeneous particles, each with parameters that vary within a specific range. Mathematically, one can accurately describe the entire granular material using continuous distribution functions that capture the behaviour of all varying parameters. However, such continuous representation is often impractical for modelling and numerical analysis. To address this, the full range of a given parameter is usually divided into several shorter discrete intervals, referred to as classes. Each class is assigned a representative value, such as the average within that interval. The material is then distributed across these classes, assuming that all particles inside the class share the same parameter values. In this way, rather than relying on continuous functions, distributions are represented as finite sets of discrete values, describing the material quantity (such as particle number, mass, or volume) within each class (Skorych et al., 2020a). Most models then track each class individually, often introducing one or more equations per class. Consequently, the number of classes in the representation is often a trade-off between the accuracy of the target distribution representation and the model's computational complexity.

Another problem is that each specific model can determine the number and composition of distributed parameters. However, at the level of the modelling system, it is necessary to preserve the ability to combine different models within a single flowsheet. And to not limit the scope of the modelling system, the number and composition of distributed parameters cannot be restricted and must be flexibly configurable. This significantly complicates the solid-state flowsheet simulation system since maintaining the consistency of distributed parameters when moving from unit to unit partially becomes the task of the modelling framework (Skorych et al., 2020a).

The challenges of solid modelling become even more profound when processes involve multiple phases, introducing solid-liquid or solid-gas interfaces. In these cases, additional interfacial phenomena, such as wetting, adhesion, and capillary effects, must be considered, which are challenging to predict and model

accurately. Overall, solid modelling requires extensive data on material properties, which can be difficult to obtain and measure with precision. Furthermore, the computational demands for these simulations are often high due to the complex, nonlinear models involved, making large-scale simulations or real-time dynamic models quite challenging to achieve.

1.3 Dynamic simulation

Regarding the flowsheet simulation, a distinction is usually made between steady-state and dynamic modelling. This classification applies both to the process simulation and to the basic models representing individual plant operations. The steady-state one assumes that all process variables, such as flow rates, temperatures and compositions, remain fixed during the simulation (Luyben, 1990). Such a model provides a snapshot of the process at equilibrium or given operating conditions. One of the basic principles here is that material that enters or is created in a process leaves it instantly, i.e. there is no accumulation of material within the process (Dimian et al., 2014a). Steady-state models tend to be simpler and more computationally efficient. However, they cannot capture transient behaviours such as start-up, shutdown or responses to disturbances, which limits their applicability in scenarios where process conditions change significantly over time.

In contrast, dynamic flowsheet modelling accounts for the time evolution of process variables (Dimian et al., 2014b) in response to the changes in input and current state of the system. These changes can have various sources, such as influences of control systems, operators, wear of critical components, alterations in raw material properties, and many more (Asbjörnsson et al., 2022). Thus, dynamic modelling makes it possible to describe not only the final state of a process but also how this state was reached (Luyben, 1990). This approach is essential for understanding transient behaviour, analysing system stability, and developing advanced optimisation and control strategies (Marquardt, 1996; Asbjörnsson, 2015; Dosta et al., 2020; Asbjörnsson et al., 2022).

Although dynamic flowsheet modelling offers advantages in capturing the behaviour of complex systems where changes over time are critical, it is inherently more complex and typically much more challenging to implement (Barton and Pantelides, 1994). Dynamic models require advanced numerical methods (Ponton, 1983; Pantelides and Barton, 1993; Borchardt and Michael, 1996; Dosta et al., 2010) and significant computational resources to solve accurately, especially for processes with many interconnected unit operations. In addition, fitting and validating dynamic models requires detailed input data that are often difficult to obtain experimentally (Montañés et al., 2017; Asprion et al., 2022). The balance between model accuracy and computational feasibility is also a significant

challenge (Laganier, 1996), as highly detailed models can be computationally disadvantageous, while overly simplified ones risk missing critical dynamics.

Many unit operations in solids process engineering, such as conveyors, bunkers or reactors, substantially impact the transient behaviour of the overall plant, making their dynamic modelling critical for applying flowsheet simulations (Gleiss et al., 2017; Metta et al., 2019; Skorych et al., 2020a). However, not all unit operations exhibit significant transient behaviour. Some operations, such as screens or specific mills with small holdup masses, can be effectively treated as steady-state even in dynamic modelling environments (Marquardt, 1991; Dosta et al., 2020), simplifying the model complexity without significant loss of accuracy. In practice, steady-state and dynamic modelling often complement each other (Dosta et al., 2010), exploiting the simplicity of the former and accounting for critical dynamics with the latter.

Overall, dynamic flowsheet modelling of solids is essential for developing robust, flexible, and efficient processes in various industries. However, the growing demand for dynamic models and the need to implement them in flowsheet simulation systems have been discussed for several decades (Marquardt, 1996; Asprion et al., 2022). Despite this, the task still remains challenging due to high computational and algorithmic demands.

1.4 State of the art

The development of the flowsheeting tools from simple steady-state models to dynamic ones, as well as from the consideration of fluid phases to granular solids, can be traced by the comprehensive review publications (Hlaváček, 1977; Motard et al., 1975; Rosen, 1980; Perkins et al., 1982; Marquardt, 1991, 1996; Moe and Hertzberg, 1994; Werther et al., 2004, 2011; Hartge et al., 2006; Merchan et al., 2016; Dosta et al., 2020). Nowadays, flowsheet simulation is a well-established methodology with a long history in process engineering, particularly in the chemical industry, which deals with fluid materials (Biegler et al., 1997; Luyben, 2002; Towler and Sinnott, 2013; Dimian et al., 2014c). There are numerous reviews in the literature of the available process simulators and related approaches used for specific applications, such as chemical (Ponce-Ortega and Hernández-Pérez, 2019) and petrochemical (Libing et al., 2021) processes, food processing (Ureta and Salvadori, 2023), biopharmaceutical production (Malinov et al., 2024), or biomass gasification (Inayat et al., 2020).

Currently, there are many commercial and open-source software packages available for flowsheet modelling, including but not limited to:

- Apros (Fortum and VTT Technical Research Centre of Finland Ltd., 2024) is a dynamic process simulation software for nuclear and thermal power plant applications (Alobaid et al., 2015; Szógrádi et al., 2020).
- Aspen Plus (AspenTech, 2024a) is commonly used in the chemical, oil, gas, and related industries (Guan et al., 2012; Le et al., 2020; Moure Abelenda et al., 2023) and has limited support for steady-state solid phase operations (Al-Malah, 2016a; Hu et al., 2017).
- Aspen Plus Dynamics (AspenTech, 2024b) is an extension of the Aspen Plus designed for modelling dynamic processes, widely utilised in the chemical and petrochemical industries for simulating fluid processes (Luyben, 2015; Al-Malah, 2016b; Taqvi et al., 2016; Sarath Yadav et al., 2022).
- AVEVA PRO/II Simulation (AVEVA Group Limited, 2024a) and AVEVA Dynamic Simulation (AVEVA Group Limited, 2024b) are steady-state and dynamic simulators for process design improvement, plant optimisation, operational analysis, and operator training in chemical, petrochemical, refining, pharmaceutical, and polymer industries (Farina et al., 2016; Talero and Kansha, 2022).
- ChemCAD (Chemstations, 2024) is process simulation software used in the chemical, fuel, pharmaceutical, and food industries (Otte et al., 2016; Petrescu et al., 2024). Has limited support for the solid phase.
- CHEMPRO (EPCON Software, 2024) is a chemical process engineering suite for simulating and optimising liquid and gas processes (Nelson et al., 2018).
- COCO Simulator (AmsterCHEM, 2024) is a free, non-commercial, steady-state flowsheet modelling environment for chemical processes (Zalazar-Garcia et al., 2022; Alqaheem and Alobaid, 2024).
- DESIGN II (WinSim Inc., 2024) is a process simulation software for steady-state modelling of chemical and hydrocarbon processes (Wang et al., 2003; Giardinella et al., 2022).
- DWSIM (DWSIM, 2024) is an open-source steady-state and dynamic process simulation tool for the design, thermodynamic modelling, and optimisation of chemical processes (Buitrago et al., 2017; Sreemahadevan et al., 2024).
- Fives ProSim (Fives ProSim S.A.S., 2024) is a family of tools for steady-state and dynamic process simulation, optimisation and thermodynamic calculations of chemical processes in fluids (Floquet et al., 2009; Ramalingam et al., 2012).
- gPROMS FormulatedProducts (Siemens, 2024a) is part of an equation-oriented general-purpose modelling environment gPROMS (Siemens, 2024b), specially designed to investigate and optimise solids processes

with a focus on the pharmaceutical industry (Barton and Pantelides, 1994; Boukouvala et al., 2012; Pinto et al., 2014; Metta et al., 2019; Paolello et al., 2023). Does not support extended solid material description with interdependent distributed parameters.

- Honeywell UniSim Design Suite (Honeywell International Inc., 2024) is designed as a tool for steady-state and dynamic modelling, process design, optimisation, decision support and operator training in chemical and petrochemical industries (Oravec et al., 2017; Viecco et al., 2023).
- HSC Sim (Metso, 2024) is a spreadsheet-based process simulation tool for chemical, thermodynamic, metallurgical, and mineral-processing calculations in steady-state and dynamic modes without detailed representation of the solid phase (Hamuyuni et al., 2021; Larssen et al., 2024).
- Integrated Extraction Simulator (Orica Limited, 2024) is a cloud-based tool specially developed for steady-state modelling and simulation in the minerals industry (Rocha et al., 2022; Mavhungu et al., 2024).
- JKSimMet (JKTech Pty Ltd., 2020) is a software for the steady-state simulation, design, and analysis of comminution and classification circuits in mineral processing operations (Zhang, 2016; Faramarzi et al., 2018).
- METSIM (METSIM International, LLC, 2024) is a steady-state and dynamic process simulation system for modelling and optimising chemical and metallurgical processes (Yahya et al., 2020; Y. Wang et al., 2021).
- OpenModelica (OpenModelica, 2024) is an equation-oriented general-purpose simulator that, among other things, allows process simulation (Nayak et al., 2019; Anjum et al., 2020).
- ProMoT (Kröner et al., 1990; Tränkle et al., 2000) is an object-oriented and equation-based process modelling tool mainly applied to chemical engineering problems (Mangold et al., 2004; Waschler et al., 2006).
- SuperPro Designer (Intelligen, Inc., 2024) concentrates on process optimisation and scheduling in pharmaceutical, special chemical, and food processing industries. Despite having a limited set of solids-involving unit operations, it lacks depicting distributed properties of granular materials (Petrides et al., 2014; Harrison et al., 2015; Rakicka-Pustułka et al., 2020).
- Symmetry (SLB, 2024) is a process simulation software for modelling fluids in the chemical, oil, and gas industries (Azhari et al., 2023; Othman et al., 2024).
- Also, various developments based on general-purpose tools like Excel, MATLAB or MATLAB-Simulink are often used for flowsheet simulations (Ali and Idriss, 2010; Gonzalez-Bustamante et al., 2007; Asbjörnsson et al., 2013; Muthukrishnan and Al Matroushi, 2018; De Carvalho et al., 2024).

Thus, despite this variety, it is easy to see that the application scope of most flowsheeting software is limited to a particular industry. Mostly, it is the chemical, gas, and oil industries, where only liquids and gases are considered. Those few simulation packages that can deal with solids are either also tailored to a specific application (mainly mining), have a limited set of unit operations, or have a simplified consideration of the solid phase without distributed parameters of the granular material.

New technologies based on advanced metaheuristic techniques, big-data analysis and artificial intelligence have rapidly developed in recent years. They are also gradually making their way into the process modelling industry, opening up new opportunities and offering fundamentally new solutions to old problems. McBride and Sundmacher (2019) gave a good overview of surrogate modelling techniques in chemical process engineering. Data-driven metamodels (Palmer and Realff, 2002), surrogate models (Nentwich and Engell, 2016; Triantafyllou et al., 2024), and deep learning methods (Agarwal et al., 2021; Zapf and Wallek, 2022; Alauddin et al., 2023) have demonstrated their potential in computational performance improvement, process and design optimisation, fault detection and classification, model generalisation, and predictive modelling. The application of artificial intelligence in particle technology generally (Thon et al., 2024) and the integration of metaheuristic algorithms (Ponce-Ortega and Hernández-Pérez, 2019) have shown promising results in improving the accuracy and efficiency of solving flowsheet modelling tasks. Coupling flowsheet simulation with different advanced methods, such as Monte Carlo stochastic techniques (Kotalczyk and Kruis, 2018), discrete element method modelling (Sen et al., 2013), or artificial neural networks (Dosta and Chan, 2022), demonstrated great potential for increasing the accuracy and expanding the scope of applicability of process simulations. Data-driven and machine-learning methods have been proposed to identify and troubleshoot convergence issues in highly interconnected systems (Ludl et al., 2022; Qu et al., 2024).

However, despite all these advancements, there is a lack of generally applicable flowsheet simulation tools capable of comprehensively considering the peculiarities of bulk materials and modelling transient processes involving them. It was this observation that prompted the German Research Foundation (DFG) to launch the research priority program SPP1679 “Dynamic Simulation of Interconnected Solids Processes” (TUHH, 2021) in 2013. The main goal was to consolidate knowledge in the area of flowsheet modelling of solids in the form of new methods, numerical tools, and dynamic process models of various processing equipment. All the new developments should have been integrated into a novel open-source software Dyssol (Skorych, 2024), presented in this work. The