Abstract

The doctoral research aims to provide a numerical platform for describing precipitation processes. Developed within the European project SEArcularMINE, the thesis aims to create models for the fundamental and applied study of magnesium hydroxide precipitation. Ultimately, these models are used in a predictive way for the development of a Magnesium Crystals-Granulometry-Controlled-Reactor (Mg-CGCR) on a pilot scale. This document provides a brief introduction to the work, a summary of thesis activities, and the main results obtained.

The research focuses on the description of magnesium hydroxide precipitation, necessary for a circular economy-based process. However, in terms of fundamental research, its precipitation is poorly characterized. Therefore, a complete description and understanding of the interference between the various involved phenomena are necessary. The identification of precipitation kinetics is achieved through multivariate regression of experimental data, guided by artificial intelligence. The data were collected using a T-mixer. The datasets aim to study two effects in the T-mixer: (i) the influence of concentration on particle sizes produced; (ii) the influence of flow rate on particle sizes produced. The first dataset was used to identify the kinetics, while the second for validation. Subsequently, the model's predictive capacity was tested on a third dataset obtained through a Y-mixer. Fluid dynamics in the Y-mixer were described through Computational Fluid Dynamics (CFD) simulations. The combination of computational simulations and kinetic parameters identified through the T-mixer predicted particle size distribution trend, compared to experimental data. The precipitation framework was further extended to simulate spatially distributed (3D) systems using the open-source software OpenFOAM. The numerical framework played a pivotal role in guiding the optimization of a first-generation Mg-CGCR prototype. A reliable numerical framework's definition involves describing all the main phenomena and their mutual interference. To adequately model the process, successive validations were conducted to ensure that all considered aspects were well-described.

A preliminary study of the flow field and turbulence inside square and circular cross-section T-mixers (with a hydraulic diameter, D_h, of 3 mm and a mixing channel length of $50D_\mathrm{h}$) was performed employing an innovative image analysis technique. This analysis validated the turbulence model and indirectly the flow field. Experimentally, macro-mixing was studied using the

dilution of two non-reactive chemical species (water with dyes, yellow in one arm and blue in the other). The pictures were analyzed, and a coefficient of variation along the mixing channel was calculated. Micro-mixing was assessed using two reactive species (hydrochloric acid and sodium hydroxide) with a change in colour indicating the pH variation due to the chemical reaction. A 3D model was implemented in OpenFOAM. The study of turbulence in these systems revealed a high intensity of local turbulence that is underestimated by common turbulence models. Therefore, local turbulence was increased by tuning the turbulence model coefficients. This tuning accurately described both macro and micro-mixing phenomena. For the description and understanding of precipitation, a simplified 1D model was developed. This model was used for determining the magnesium hydroxide precipitation kinetics. The model accounts for chemical species reactions resulting from micro-mixing (using $k-\varepsilon$ profiles from the preliminary fluid dynamics study) and ionic activity to correct non-ideal behaviour at high concentrations. The model generates supersaturation, used to calculate primary nucleation and growth rates, and uses aggregation kernels for hydrodynamic and Brownian collisions. Kinetic parameters were determined through multivariate optimisation with an innovative AI-guided method. Model results were compared with experimental data obtained in the T-mixer for each investigated concentration. The validated set of kinetic parameters was then used to describe other experimental datasets, including a different T-mixer dataset and a Y-mixer dataset. The Y-mixer setup involved a mixer with a diameter of 1 mm and a length of 3 mm, and the computational fluid dynamics simulations accurately predicted the system's particle size distribution. The model's ability to predict the system's behaviour was tested in 3D configurations to consider their complex fluid dynamics. A 3D model was implemented and successfully tested in OpenFOAM, serving as a support tool for the Mg-CGCR prototype's optimization.

In summary, the key points of the research activities are presented. (i) The description and validation of the flow field involved implementing a model calibrated and validated with experimental data obtained through image analysis. (ii) Implementing a simplified model allowed the extraction of a reliable set of parameters using artificial intelligence. (iii) The set of kinetics was validated and used to predict particle sizes in a Y-mixer. (iv) A 3D model was implemented and successfully tested in OpenFOAM, serving as a support tool for the Mg-CGCR prototype's optimization.