Growth rate of silica particle in a continuous reactor with Computational Fluid Dynamics and Population Balance Modelling

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Introduction

- FLOW4SOLID is a collaborative project within the framework of the ERDF program (Project portfolio INTENSE4CHEM), which aims to intensify the synthesis and manufacturing of Alumino-Silicate materials (e.g. Zeolites). The project globally looks on the whole process from synthesis to crystal formation, crystal growth and drying (figure 1). Three research units are involved in this project: 1- Certech defines the optimal conditions (concentration, pH, temperature, ...) of synthesis and formation of first crystals; 2- At ULiège – group PEPs, the study focuses on the innovative design of a continuous reactor in which the crystal growth will occurred; 3- The operations related to the shaping and drying of the crystals are intensified at UCL - IMAP.
- Currently, zeolite production is based on hydrothermal processes performed under controlled condition (temperature and pressure). The crystal properties are adjusted by adjusting crystallization parameters including reaction duration and reactant concentrations [1].
- Crystallization process of zeolites is most often carried out in batch stirred tank reactors which causes many limitations in term of productivity and operating cost. Thus, a way to overcome these drawbacks, is to switch the process from batch to continuous which has several advantages including more reliable reaction control, saving energy and reducing the impact on the environment.
- In the present work, the growth of silica nanoparticles (as a particle model for zeolites) is analyzed in a continuous 2D reactor with the objective of optimizing the process via the coupling between all major phenomena occurring within the reactor.

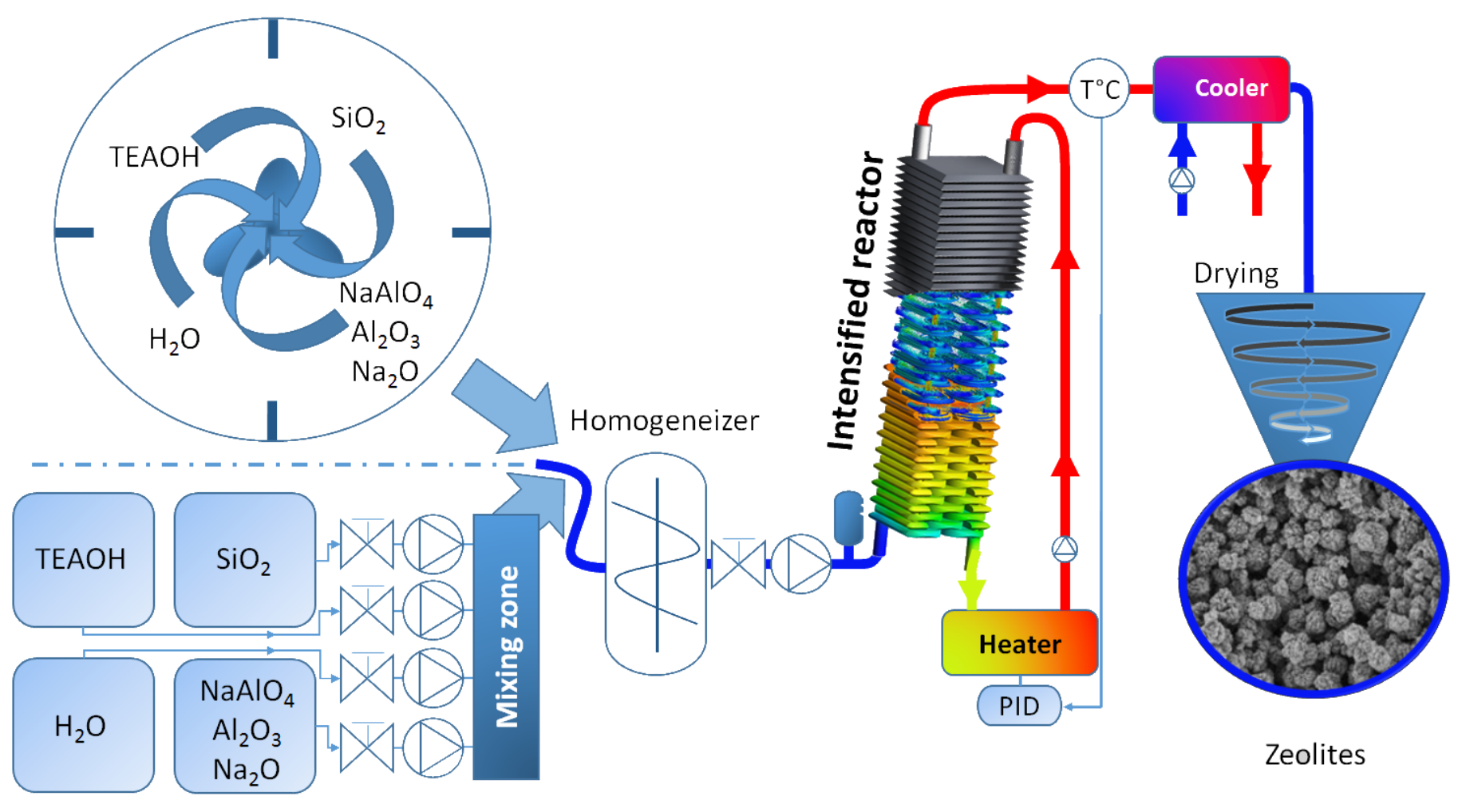

Figure 1 – Flow4Solid complete project

Process Description & Case Study

PROCESS DESCRIPTION

- A typical crystallization process consists in two steps: the first step is nucleation, during which a crystalline phase (i.e. nuclei) is formed from a supersaturated suspension of dissolved molecules. In the second step, the crystal size in the suspension is further increased to a desired value (i.e. growth step) [2]. Although it is not possible to completely separate the nucleation and growth processes, it is possible to choose operating conditions such that one of the reacting steps is favoured, such that the 2 steps may be successively conducted in two distinct reactors; in the first reactor crystals are produced (figure 2-a) and then they are supplied to the second reactor for the further growth (figure 2-b).
- Particle Size Distribution (PSD) is an important property of crystallization quality and is aimed to be controlled in this process. Therefore, Population Balance Model (PBM) is applied to predict the PSD subject to particle growth and various hydrodynamic conditions found in continuous reactor.

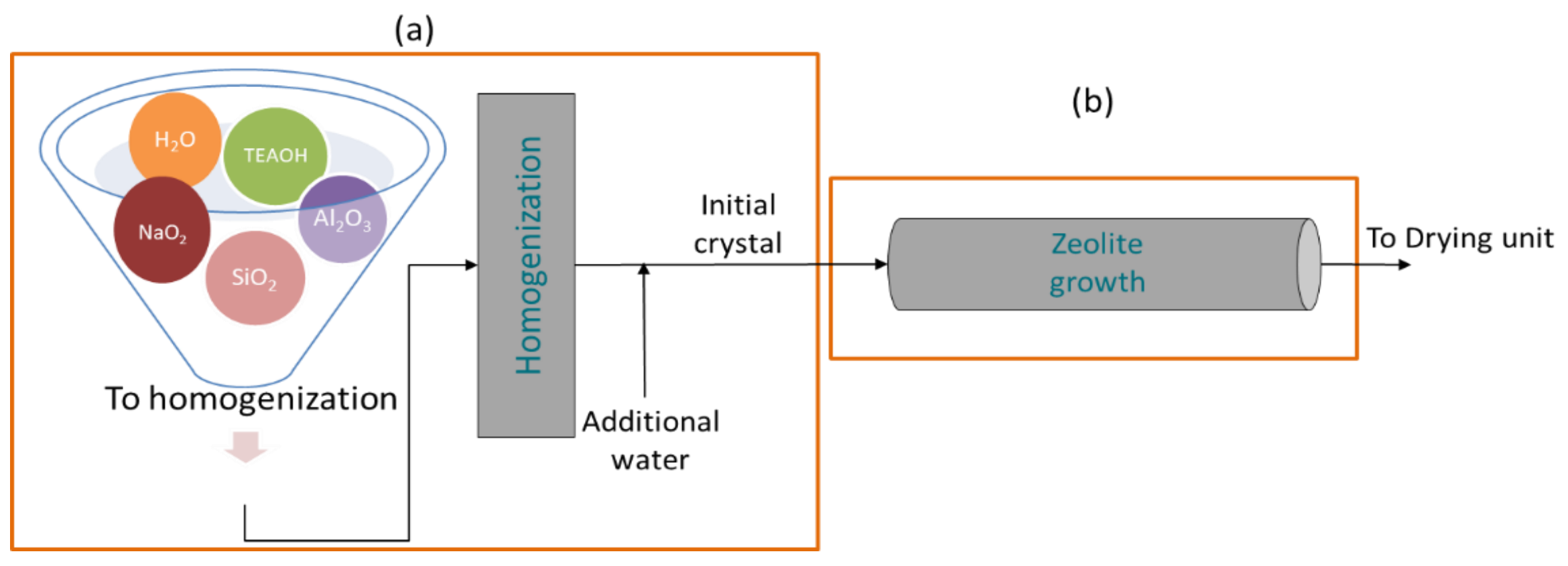

Figure 2 – Schematic diagram of crystallization process: a) first step – synthesis and crystal formation; b) second step – crystal growth

CASE STUDY

- A continuous 2D reactor has been modelled in this study. Amorphous silica particles were selected as reference particles and were assumed to be spherical, initially monodisperse and nanometric. The growth rate was kept constant while the hydrodynamics were simulated using Eulerian-Mixture model.
- All the simulations presented in this work were carried out on the commercial CFD software package FLUENT 19.2 (Ansys Inc., US) due to his appropriately for the complex physics involved in a PBM-CFD.

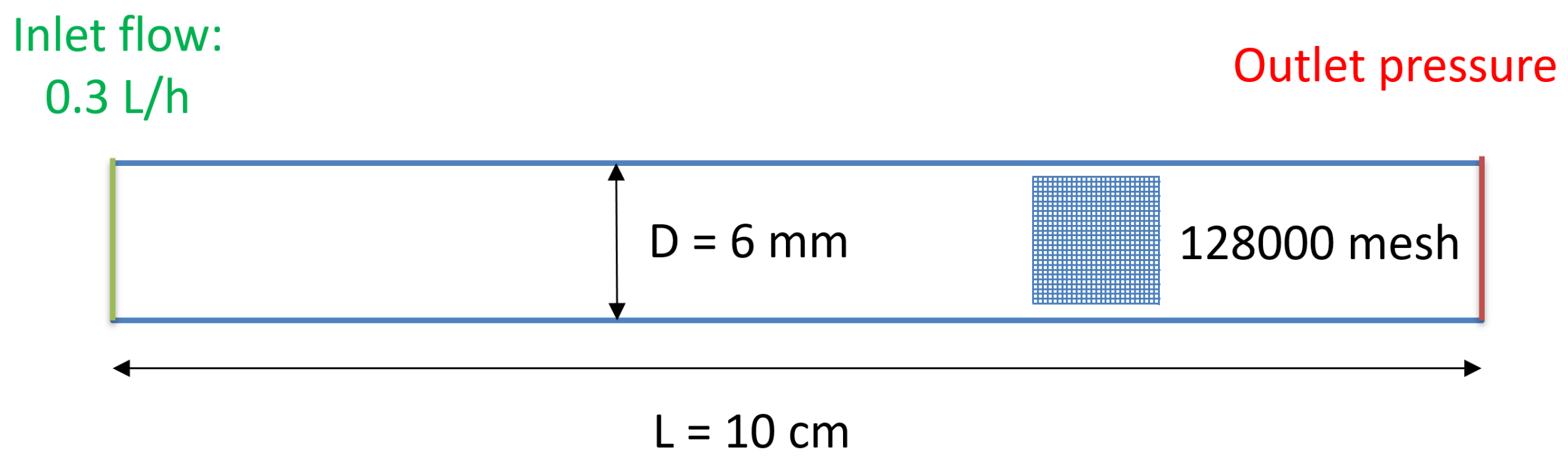

Figure 3 – Numerical Configuration used as 2D reactor

RESULTS
I. PBM validation: Plug Flow

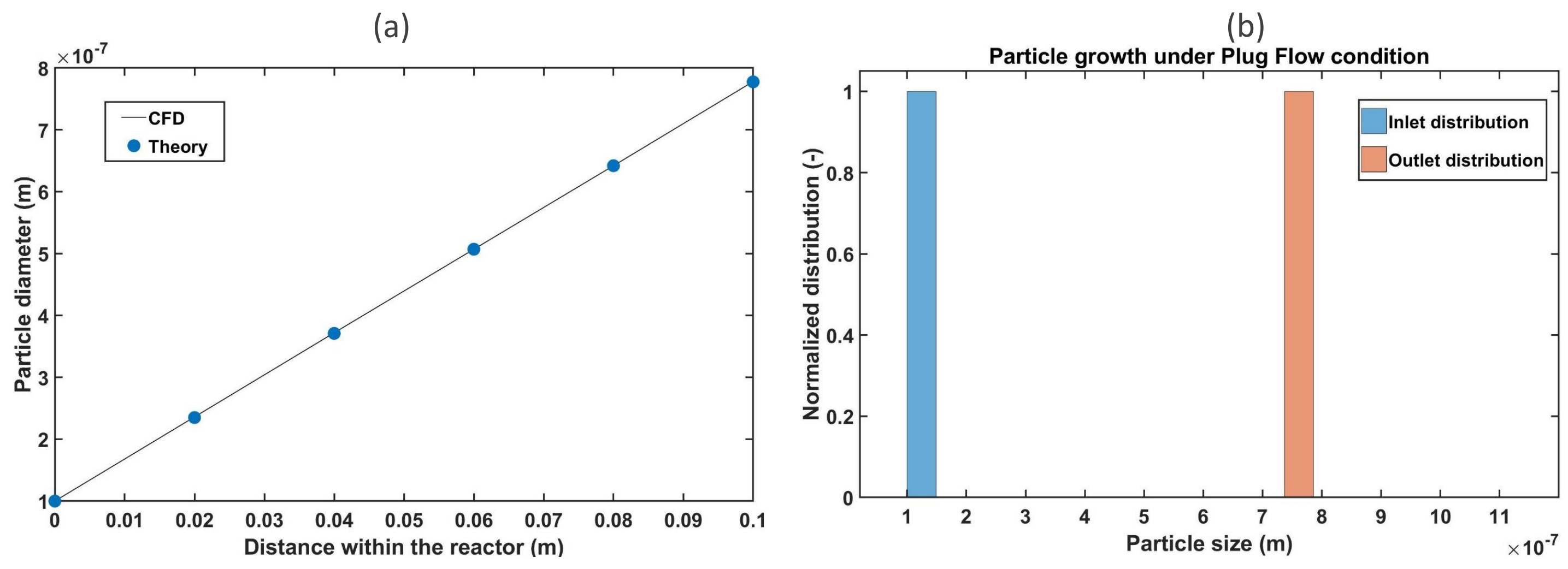

Figure 4 – Numerical result validation: a) numerical and theoretical comparison; b) Inlet outlet distribution for a plug flow regime under plug flow regime for monodisperse distribution

- To make sure that the solution of the PBM is accurate enough, the algorithm validation of the PBM was primarily carried out.
- Method of classes is firstly applied due to the facility of obtaining the real distribution. However, the use of this method led to numerical diffusion and prevented to obtain accurate results.
- Standard Method of Moments was then applied, and the results were compared to the theoretical solution under the assumption of Plug Flow. It was shown that the numerical simulation agreed with the theoretical results (figure 4-a).
- Under plug flow condition by having monodisperse distribution at inlet and the constant growth rate within the reactor, all particles must grow in the same way and they must all have the same size at outlet.
- Numerical results validated this concept and showed that all particles have indeed the same size at outlet (figure 4-b) which can be considered as an additional technique to validate the numerical simulation.

References

[1] Z. Liu, J. Zhu, C. Peng, T. Wakihara and T. Okubo, React. Chem. Eng., Vol. 4, 1699-1720 (2019).
[2] T. Wakihara and T. Okubo, Chemistry Letters Vol.34 (3), 276-281 (2005).

RESULTS
II. Hydrodynamic: Laminar Flow

- The influence of hydrodynamics on the PSD was investigated in laminar flow conditions and different inlet flow rates (i.e. different average residence times).
- The final PSD for two different residence times under the laminar flow conditions were simulated and compared (Figure 5). Logically the average particle diameter got bigger and the PSD became larger when the residence time increased.

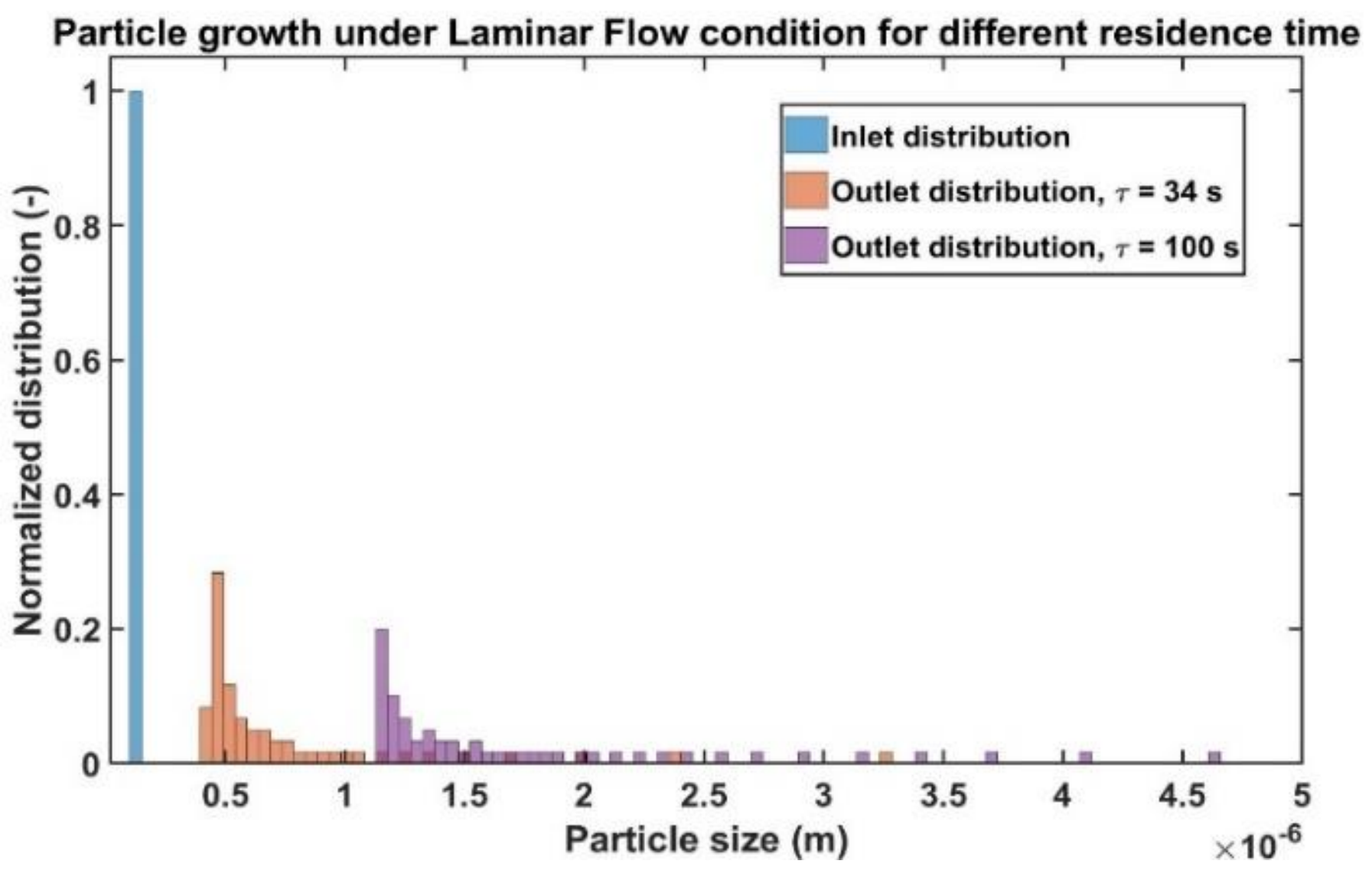

Figure 5 – Particle size distribution under laminar flow condition and after different residence time

RESULTS
III. Rheology : Non-Newtonian viscosity

- Experiments showed that the viscosity of silica/water suspension varies depending on the shear rate (i.e. non-Newtonian pseudo-plastic). The rheology of the suspension can be expressed according to the Ostwald-de Waele law:

$$\eta = k \cdot \dot{\gamma}^{n-1}$$

- Where η is the viscosity (Pa.s), k consistency factor (Pa.s^n), $\dot{\gamma}$ shear rate (s^{-1}) and n flow behaviour index (-).

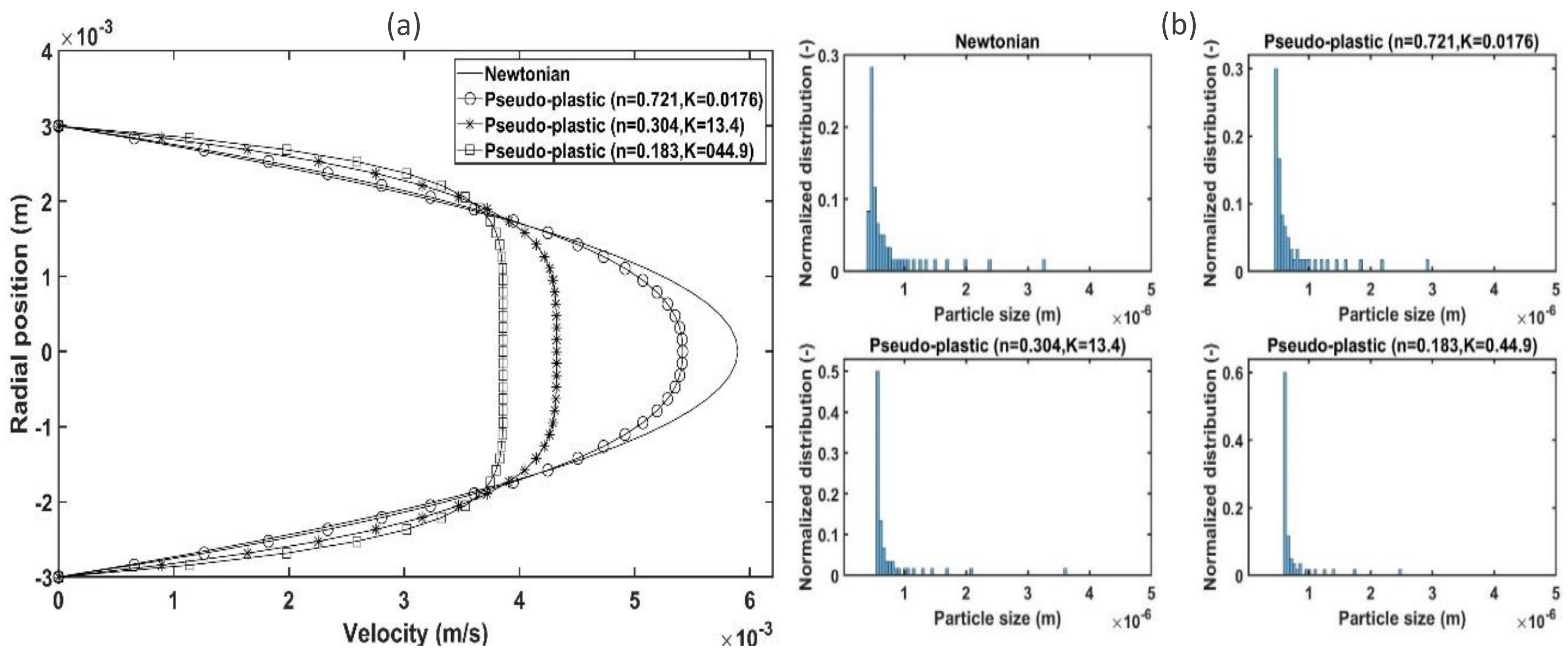

Figure 6 – The effect of pseudo-plastic viscosity: a) on velocity profile; b) on particle size distribution

- Figure 6 illustrates the effect of pseudo-plastic viscosity on velocity profile (a) as well as on PSD (b).
- It is known that the velocity profile becomes flatter when the pseudo plasticity increased (figure 6-a). The flatter flow profile caused a more uniform radial residence time distribution in the reactor resulting in a narrower PSD (figure 6-b).

CONCLUSIONS & PROSPECTS

CONCLUSIONS

- A continuous crystallization process in a 2D configuration was simulated using CFD−PBE model which enables us to successfully follow the evolution of particle size through the reactor under different conditions.
- Numerical investigations were carried out and results were compared to theoretical values.
- The effect of non-Newtonian viscosity was investigated and its impact on the velocity profile and on PSD was analyzed and presented.
- Despite the need for more details on process, this important feature helps to understand the particle distribution and growth as a function of residence time and viscosity.

PROSPECTS

As future investigations, it is planned to account for more phenomena in numerical simulations to complete the model and improve the results:

- Apply crystal growth depending on supersaturation level and temperature.
- Establish the coupling between particle growth, hydrodynamics and fluid rheological behavior.

Acknowledgement

The authors gratefully acknowledge the support of Wallonia region and European Regional Development Fund (ERDF).

GEPROC, Mons - Belgium, November 7, 2019