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Simulation for scale-up of a confined jet mixer for continuous hydrothermal flow synthesis of nanomaterials

Cai Y. Ma[a], Jing J. Liu[b], Yang Zhang[b], Xue Z. Wang[a,b,*]

[a] Institute of Particle Science and Engineering, School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT, United Kingdom
[b] School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou City 510640, PR China

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ABSTRACT

Reactor performance of confined jet mixers for continuous hydrothermal flow synthesis of nanomaterials is investigated for the purpose of scale-up from laboratory scale to pilot-plant scale. Computational fluid dynamics (CFD) models were applied to simulate hydrothermal fluid flow, mixing and heat transfer behaviours in the reactors at different volumetric scale-up ratios (up to 26 times). The distributions of flow and heat transfer variables were obtained using ANSYS Fluent with the tracer concentration profiles being simulated via solving the species equations. The predicted temperature distributions under various volumetric scale-up ratios were compared with the available experimental data, and good agreements reached. The mixing between supercritical water jet and precursor stream with different scale-up ratios was examined in detail to identify the effect of scale-up ratios on hydrodynamic and thermodynamic features. The findings indicate that slightly weaker mixing was observed at the pilot plant scales, but the momentum dominated turbulent flow in the reactors and the same order of magnitude of mixing levels at both laboratory and pilot plant size scales could lead to similar quality nanoparticles to be manufactured under the investigated volumetric scale-up ratios and operating conditions, which is supported by experimental observation from literature.

1. Introduction

Development of nanotechnology can be divided into several levels: materials, devices and systems. Therefore nanomaterials (particles <100 nm) are at the leading edge of the emerging field of nanotechnology. Their unique size-dependent properties make these materials superior and indispensable in many technological applications. The inability to successful scale-up and manufacture of high quality nanomaterials in a consistent way at commercial scales represents a major challenge. The current study investigates a promising nanomaterial process, the continuous hydrothermal flow synthesis (CHFS) process, using computational fluid dynamics (CFD) as a tool for the scale-up of the CHFS system.

Supercritical water hydrothermal synthesis has been used to produce numerous metal oxides, in both batch and continuous modes [1–6], with wide applications [7]. Continuous hydrothermal flow synthesis methods were developed to overcome the limitations of hydrothermal batch reactions [2] and able to produce very fine nanoparticles for a number of functional materials [8–14] at laboratory size scales. However, the milligram- or gram-scale laboratory systems for nanomaterial manufacture cannot meet the requirement of large amount of nanomaterials for the increasing commercial demand. This necessitates the demanding research on the optimisation and scale-up of nanomaterial manufacturing processes.

In scaling-up a process from the laboratory to pilot plant scale (and industrial scale), the development of a successful scale-up strategy based on the in-depth understanding of the hydrodynamic and thermodynamic features during reactor design and optimisation processes is critically important, but often difficult and costly to be achieved through only experiments for alternative reactor designs due to the huge number of experiments that have to be conducted and the extreme conditions of the supercritical experiments. Based on the 24 principles of green engineering and green chemistry [15], some key attributes were proposed by Gruar et al. [11] for the sustainable scale-up of nanoparticle synthesis processes and also applied to the CHFS system at pilot plant scale. Other organisations have also developed pilot-scale or larger CHFS plants which can manufacture nanoparticle product with a capacity of over 1 kg per hour [16]. Hanwha Chemical Corporation, Republic of Korea, has not only a pilot plant CHFS system, but also the first full

* Corresponding author at: School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT, UK. Tel.: +44 113 343 2427; fax: +44 113 343 2384.
E-mail address: x.z.wang@leeds.ac.uk (X.Z. Wang).

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industrial-scale CHFS plant in the world, which can have a capacity of over 200 tonnes per year [16]. However, to the best of our knowledge, relatively few results obtained from these processes are available in the general academic literature, not to mention the detailed investigation results of optimisation, control and scale-up of the CHFS reactors. Our previous studies of counter-current and confined jet reactors at laboratory size scale confirmed that the faster mixing in the confined jet mixer produced better reactor performance including more uniform size distribution, which demonstrated the advanced capacity of CFD technologies in the successful investigation of reactor design and optimisation of the CHFS processes [17]. This also led to the selection of the confined jet reactor for the scale-up process and further research on the effect of scale-up factors on reactor performance in this study.

CFD modelling is a useful tool to study the potential effects of the scale-up factors at different reactor sizes and to evaluate alternative design as the method can provide the hydrodynamic/mixing and heat transfer behaviours in reactors under various operating conditions and volumetric scale-up ratios (defined as the ratio of the volumetric inlet flowrates between the larger and smaller reactors). The method has been used to study supercritical water oxidation processes by some researchers [18–23]. The application of CFD modelling to supercritical water hydrothermal synthesis for the production of nanoparticles using impinging jet reactors is still limited in literature with very few experimental data available for model verification and comparison. Lester et al. [7] used CFD technique to simulate the velocity distributions of a nozzle reactor. The simulation was based on a pseudo supercritical water reactor with methanol and sucrose being used to represent supercritical water and metal salt. Aimable et al. [6] carried out numerical simulation in the mixing zone of a reactor with X-shaped geometry and obtained the distributions of temperature and velocity in this zone but without comparisons with measurements. Kawasaki et al. [24] studied continuous supercritical hydrothermal process in a T-shaped mixer for the experimental synthesis of NiO nanoparticles and CFD simulation. The simulation was performed to understand the mixing behaviour in the mixer and its heating rate with no experimental data for comparisons. Sierra-Pallares et al. [23] attempted to quantify the mixing efficiency in turbulent supercritical water hydrothermal reactors including a T-shaped mixer and a counter-current reactor, but without comparison to experimental data. Further study was carried out to develop a model for the precipitation of β-carotene using an antisolvent process with supercritical carbon dioxide [25]. Demoisson et al. [22] performed CFD simulations for the design of a reactor operating in supercritical water conditions with some samples of synthesised nanomaterials. A representative mixing temperature of reaction medium was obtained with a thermocouples located close to the reaction zone. The study then compared the measured mixing temperature of individual experimental runs with the corresponding simulation temperature in the reaction zone. Ma et al. [17] numerically investigated the hydrodynamic and thermal behaviours in a confined jet mixer and a counter-current reactor under supercritical water conditions with the predicted temperature distributions for both reactors being compared with the experimentally measured data. Anti-solvent precipitation processes were simulated for the production of paracetamol crystals from ethanol solution using supercritical carbon dioxide as an anti-solvent [26]. The simulated distributions of velocity, temperature and species concentration were not compared with experimental data which may be not available due to extremely difficult measurement conditions. It was suggested that the best and safe scale-up method for the investigated processes using supercritical CO_2 is to use high Reynolds number (hence, well developed turbulent) flows in the reactors. To the best of our knowledge, there is no report in the literature that the scale-up processes of a reactor with confined jet configurations under various supercritical water conditions and scale-up ratios were investigated in detail.

In this paper, CFD models were developed and applied to predict the process behaviour in confined jet reactors at both the laboratory and pilot-plant scales under various volumetric scale-up ratios. The predictions of flow, mixing and temperature profiles in the reactors were carried out using the ANSYS Fluent [27] with the temperature profiles being compared with the available experimental data [11,12]. The simulated results under various scale-up ratios were further examined to identify the effect of reactor scale-up on its performance.

2. Experimental description

Fig. 1 shows the schematic drawing of a CHFS confined jet mixer system [11,12,17] used for temperature profiling and also nanoparticle production experiments. Pump 1 was used to pump

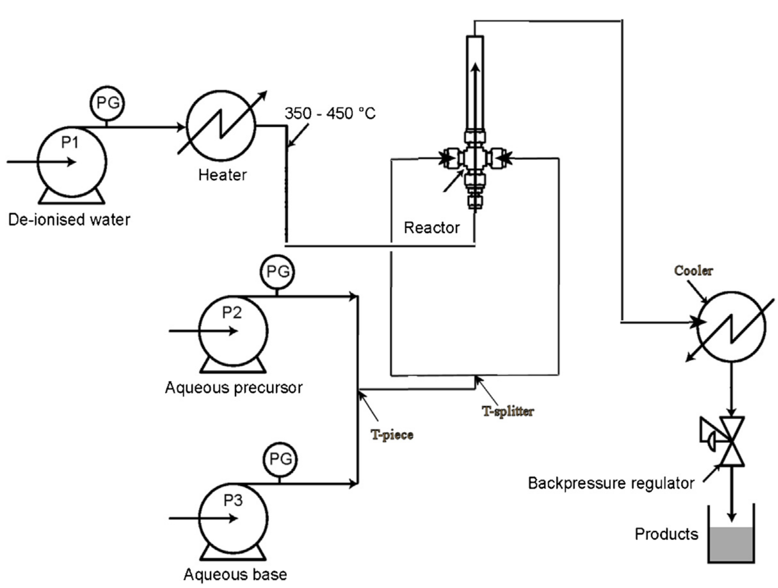

Fig. 1. Flow diagram of a confined jet reactor in a CHFS system [11,12,17].