Separation and Purification Technology 176 (2017) 231–242

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Filtration of aqueous colloidal ceria slurries using fibrous filters – An experimental and simulation study

Vahid Rastegar [a,b], Goodarz Ahmadi [b,c], S.V. Babu [b,d,*]

[a] Material Science & Engineering, School of Engineering, Clarkson University, Potsdam, NY 13699, USA
[b] Center for Advanced Materials Processing, Clarkson University, Potsdam, NY 13699, USA
[c] Department of Mechanical and Aeronautical Engineering, Clarkson University, Potsdam, NY 13699, USA
[d] Department of Chemical and Biomolecular Engineering, Clarkson University, Potsdam, NY 13699, USA

ARTICLE INFO

Article history:
Received 27 April 2016
Received in revised form 29 November 2016
Accepted 12 December 2016
Available online 18 December 2016

Keywords:
ANSYS-FLUENT
Fibrous filters
Fiber efficiency
Chemical mechanical polishing
Colloidal ceria dispersion

ABSTRACT

A combined experimental and computational study of fibrous filters for removal of large hydrosols, essential to minimize defects during chemical mechanical polishing, was performed. Dilute aqueous suspensions of colloidal ceria particles, of known size distribution, were filtered and the filter efficiencies were measured for different particle sizes and pH, then converted to single fiber efficiencies. The particle size distributions were also measured for the influent and effluent streams.

In a series of numerical simulations, the Navier-Stokes equation was solved for a single fiber using the ANSYS-FLUENT computational fluid dynamics commercial package. For dilute suspensions, the motion of the dispersed particles in the size range of 35–600 nm was tracked in the Lagrangian reference frame including the effects of hydrodynamic drag, lift, gravity, hydrodynamic retardation, Brownian, van der Waals and electric double layer forces. The electric double layer and van der Waals forces were incorporated in the calculations by developing a user defined function. Particular attention was given to the effect of Brownian excitations, as well as the electric double layer and van der Waals forces - which have been neglected in many of the previous models - on the overall fiber collection efficiency for different particle sizes and charges. The simulated fiber efficiencies and particle size distributions compared very well with experimental results.

1. Introduction

Chemical mechanical polishing (CMP) is a widely used process for manufacturing integrated circuits (ICs). Abrasive particle size in the slurry is typically in the range of 30–140 nm and the presence of larger abrasive particles can cause defects (such as pits and microscratches) and yield losses. Selective removal of large defect causing particles without affecting the polishing performances is one of the goals of effective slurry filtration. Fibrous filters can be classified as woven and nonwoven filters. Nonwoven filters are more commonly used in filtration technology [1,2]. Fibrous filters have long been showed to be capable of trapping aerosols successfully [3,4]. More recently fibrous filters were used for filtering solid particles in aqueous suspensions [5–7]. In most filtration applications such as water treatment [8,9], the goal is separating the particulate phase with the maximum collection efficiency. For filtration of slurry for CMP applications, however, the filter media should be designed in such a way that the larger particles are selectively removed and the particle size distribution becomes narrower around the desired size. Many studies on developing mathematical formulations for predicting the performance of fibrous filters for airflows were reported in the literature [10–19]. For aqueous dispersions, however, the amount of works is comparatively limited [20–23]. In particular, the effects of electric double layer (EDL) and/or van der Waals (vdW) forces were neglected in many of the existing models. O'Melia and Stumn [24], Tien et al. [25–34] and Yao et al. [35,36] reported their interested study on hydrosols filtration in granular collectors using particle trajectory analysis considering limiting trajectories. The approach, however, has certain limitations for estimating the rate of particle deposition [18]. In particular, inclusion of the Brownian diffusion effects is challenging owing to its stochastic nature [37]. Hence, a comprehensive Lagrangian trajectory analysis model for nano-particle filtration that covers all the relevant forces is still lacking.

* Corresponding author at: Center for Advanced Materials Processing, Clarkson University, Potsdam, NY 13699, USA.

E-mail address: babu@clarkson.edu (S.V. Babu).

http://dx.doi.org/10.1016/j.seppur.2016.12.017
1383-5866/

In general, there are two methods for two phase flow analysis in computational fluid dynamics (CFD) simulations, the Eulerian-Eulerian and the Eulerian-Lagrangian method. The Eulerian-Eulerian method treats the particle phase as a continuum similar to the fluid phase and develops and solves the corresponding equations for conservation of mass and balance of momentum for the particle phase as well as for the fluid phase [38–42]. In this approach constitutive models need to be developed for various phasic properties and the interaction forces. The Euerian-Lagrangian method treats the fluid as a continuum but considers particles as a discrete phase and tracks the pathway of individual particles [16,43–45].

Since a fibrous filter has an open structure, suspended particles are collected by coming into contact with a fiber and adhering to its surface by the surface forces. For dilute particle concentration and low fiber volume fraction, the particle collection process on a single fiber does not interfere with deposition on other fibers. Therefore in the simulations, the filtration process can be modeled as particles interacting with single fibers in their flights through the filter. There are different mechanisms for the deposition of particles on the fibers. For particles larger than a few μm, the inertial impaction and sedimentation mechanisms are dominant, while Brownian motion plays a greater role for particles smaller than 0.1 μm. For particles in the size range of 0.1–0.5 μm, the interception mechanism becomes more important as both particle inertia and Brownian diffusion are relatively weak. As a consequence, the collection efficiency of a fibrous filter has a minimum in this size range. Electrostatic and van der Waals forces can increase the collection efficiency of particles, particularly, in the size range of 0.15–0.5 μm [46,47].

In this study, using a one-way coupling assumption, the filter efficiency under various conditions was simulated. A single fiber was placed with its axis perpendicular to the fluid flow in a 2-D computational box and the flow field around the fiber was evaluated using the commercial CFD software - ANSYS-FLUENT. Once the flow field was obtained, particles with diameters in the range of 35–600 nm were introduced into the flow using the discrete phase model (DPM) in the ANSYS-FLUENT code. The trajectories were evaluated using the particle equation of motion in the Lagrangian reference frame. The computational model in the software included all hydrodynamic forces, but did not account for the surface forces. A user defined function (UDF) was developed and compiled in the ANSYS-FLUENT model using C programming language to include the hydrodynamic retardation effects and EDL and vdW forces. Effects of particle charge on particle trapping for both like charge and opposite charge surfaces were studied. Furthermore, the effects of Brownian, EDL and vdW forces on fiber collection efficiency were investigated. The single fiber simulation results were extended to the fibrous filter efficiency using the available empirical models. The simulated filter efficiencies were compared with the experiment data and good agreement was found.

2. Experimental procedure

Nonwoven fibrous filters are commonly used for the filtration of particulates in numerous applications. Fig. 1a shows a scanning electron microscopy (SEM) view of the Polypropylene fibrous filter used in the present work. The cylindrical fibers have a mean diameter of 2 μm with a surface that appears very smooth. In the present experimental study, colloidal ceria particles dispersed in deionized (DI) water with additives of Nicotinic acid and Proline were filtered. Fig. 1b shows the schematic diagram of the filtration tests. The slurry container was placed on a magnetic mixer and the slurry was continuously stirred during the filtration test using a magnetic Teflon bar to assure particle dispersity. The slurry was delivered to the filter capsule using a peristaltic pump. Filter face velocity was altered by changing the slurry flow rate. Pressure drop was measured at a given fluid flow rate using a pressure gage in which the difference between the upstream (influent) and downstream (effluent) pressure was measured. In order to evaluate the filter efficiencies, the concentration of particles before and after filtration was compared and the fraction of trapped particles was evaluated. Ultraviolet–visible (UV–vis) spectrophotometry was used to determine the concentration of ceria dispersion samples. The samples were analyzed in quartz cuvettes with an optical path length of 10 mm using a Perkin-Elmer Lambda 35 UV–vis spectrophotometer. Each sample was sonicated for twenty seconds before the UV–vis test to remove any air bubbles. Fig. 2a shows the UV–vis spectra of ceria dispersions with different ceria particle concentrations. It is seen that the extinction of light increases as the concentration of solid particles increases. Fig. 2b shows the variation of the extinction of laser light at the wavelength of 365 nm with the ceria slurry concentration. This figure was obtained using different slurry samples with known ceria concentrations. This figure clearly shows a linear relationship between the particle concentration and light extinction. Concentrations of ceria in the slurry in the filtration experiment were evaluated using the calibration curve of Fig. 2b. Filter collection efficiencies were determined from measured concentrations of feed and effluent dispersions. That is,

$$\textit{Filter efficiency} = 1 - \frac{C_{effluent}}{C_{feed}} \tag{1}$$

Zeta potential of the dispersions was altered by changing the pH. Nitric acid was used to lower the pH, while potassium hydroxide was used to increase the pH of the dispersions. This was performed in such a way that the ionic strength of the dispersions in all cases remains the same by adding salt. Zeta potential and electrical conductivity of the influents and effluents were measured using dynamic light scattering technique (Malvern Zetasizer nano ZS, 532 nm 'green') using a dip cell. Fig. 3 shows the zeta potential of the ceria slurry as a function of pH. The measured zeta potentials of these particles are very similar to those obtained earlier [48–50].

Field emission scanning electron microscopy (FE-SEM) JEOL 7400 was used to evaluate the size and morphology of filter fibers and trapped particles.

Particle size distribution of the effluents was measured using dynamic light scattering technique (Malvern Zetasizer nano ZS, 532 nm 'green').

3. Formulations

3.1. Computational domain

The computational configuration and boundary conditions are described in this section. For dilute particle concentrations the assumption of one-way coupling is used that is the particle is carried by the fluid, while the flow is not affected by the particles. The fiber volume fraction in the filter is about 20%. Under these conditions, it is assumed that the particles interact with single fiber at a time and a periodic computation cell around each fiber is considered to be adequate for analysis. Fig. 4 shows the 2-D computational cell used in the simulations that constructed using the same fiber size and porosity as the experiments fibrous filters. The fluid is assumed to flow into the domain from the top where a velocity inlet boundary condition was used. The flow leaves from bottom face under a pressure outlet boundary condition. The computational cell size used in the simulations is 4×4 μm and a space with one half of the fiber diameter was added to the downstream region to assure a roughly uniform velocity at the outlet avoiding