



# Discrete element method simulations and experiments of dry catalyst impregnation for spherical and cylindrical particles in a double cone blender



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## ARTICLE INFO

### Article history:

Received 18 August 2016

Received in revised form 3 May 2017

Accepted 11 May 2017

Available online 15 May 2017

### Keywords:

Catalyst impregnation

Discrete element method

Granular mixing

## ABSTRACT

The dry impregnation of catalyst supports is a widely used process in the preparation of heterogeneous catalysts, however there has not been a lot of work done computationally on this process. In this work, discrete element method (DEM) simulations coupled with an algorithm for the transfer of fluid to and between particles are used to study dry impregnation. We use a previously developed model, which has been further validated with geometrically equivalent experiments and the results show very good agreement. The effects of rotational speed, particle size and particle morphology are explored in order to achieve the best overall mixing and fluid content uniformity in the particle bed. We study spheres and cylinders of different sizes and aspect ratios. Axial mixing analysis and liquid distributions are used to investigate the propagation of the fluid throughout the particle bed with the goal of understanding the effect of operational and material parameters and ultimately to improve fluid content uniformity in systems with particles of different morphologies. The fluid content uniformity is characterized by the relative standard deviation (RSD) of the liquid content from all the particles in the system. Our results show that cylinders always take less time to mix than spheres of the same diameter and mixing times are also shorter for cylinders of higher aspect ratios when compared to cylinders of smaller aspect ratio. Likewise, the times to reach good fluid content uniformity are shorter for cylinders with higher aspect ratios as compared with cylinders with lower aspect ratios. We also observe that mixing time in the axial direction for both spheres and cylinders followed an exponential function of the surface area to volume ratio. We found a linear correlation between the times to achieve good axial mixing and the times to achieve good fluid content uniformity in the entire particle bed, which suggests that mixing in the axial direction controls fluid uniformity in the entire particle bed.

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## 1. Introduction

Impregnation is a crucial step in the manufacturing of heterogeneous catalysts [1–3]. Generally, the process of impregnation is performed in rotating vessels with one or more nozzles that distribute a solution of the metal precursor into the catalyst support particles. As the liquid penetrates into the dry particles, the particles absorb the fluid, and the density of the particles increases accordingly until they become saturated [4,5]. After saturation, the excess liquid forms liquid bridges between particles causing wet cohesive forces that disrupt particle flow and mixing and even form particle agglomerates [6]. All of these can adversely affect the homogeneous distribution of the metal precursor in the solid, resulting in unacceptable content variability in the final catalyst product.

While dry impregnation is a widely used technique in catalysis manufacturing, the process is very seldom studied using computer simulations. Most research in this field has been done experimentally and mostly focused on aspects of the surface chemistry of the impregnation and the metal content inside individual support particles [7–9]. There have also been some transport models but those have covered a small window of the process [2]. In particular, discrete element method (DEM) has been increasingly used to study granular flow systems [10, 11]; however, most of this work is focused on the pharmaceutical area [12–14]. The catalyst manufacturing industry uses many similar processes. Heat and liquid transfer has been studied in a rotary kiln using DEM [15,16]. However the modeling of catalyst impregnation processes using DEM remains highly unexplored. To the best of our knowledge, the only reported DEM simulation study on dry impregnation of catalysis was done by our group [17]. In our previous study, we developed a fluid transfer algorithm and implemented it into a discrete element model to simulate the catalyst impregnation process. The simulations

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were validated by experiments utilizing a geometrically identical double cone blender with a single nozzle impregnator. The effects of two process parameters (i.e. flow rate and fill level) were explored. However, several effects were left unexplored, such as the effect of rotational speed and the effect of particle size and morphology. These are crucial parameters that may influence proper mixing in the particle bed for all the particles to receive similar amounts of liquid.

Double cone blender is a standard industrial tumbling mixer and commonly used for catalyst impregnation. Mixing has been experimentally shown to not be a function of rotational speed in a double cone blender [18,19]. Using a multi-dimension double cone blender that rotates around two axes, it has been concluded that the rotation of the mixer at higher speeds around the horizontal axis resulted in a better mixing efficiency and lower mixing time [20,21]. These studies, however, were done in the absence of water in the particle bed. When liquid spraying is present, it creates moisture in the granular system leading to cohesion as a result of capillary forces, which can have a significant effect on the mixing and flow behavior of granular materials [22–24]. In all these previous studies, mixing under fluid spray as a function of rotation speed has not been previously quantified.

In this paper, we investigate the overall mixing and fluid content uniformity in the liquid-impregnated particle bed at different rotation speeds and different particle sizes and morphologies. Particle size and distribution of particle sizes have a strong influence on the random diffusive particle motion in a granular bed. To the best of our knowledge this has never been reported in the literature. The remainder of this paper is organized as follows: simulation method and model validation are described in Section 2, experimental setup is given in Section 3, and results are presented in Section 4, followed by conclusions and recommendations, which are presented in Section 5.

## 2. Simulation method

Discrete element method (DEM) has been increasingly used to study granular materials and particle systems. In the DEM, the motion of individual particles is computed according to the Newton's second law of motion. The translational and rotational motions of a solid particle are expressed by:

$$ma = \sum F_{Contact} + \sum F_{Body}, \quad (1)$$

where  $m$  and  $a$  are the mass and acceleration of a solid particle, respectively. The term  $\sum F_{Contact}$  accounts for all the normal and tangential contact forces, which are due to particle-particle or particle-boundary collisions.  $\sum F_{Body}$  denotes the sum of all body forces due to gravity.

DEM simulations in this work were performed using the EDEM commercial software package, developed by DEM Solutions, Ltd., which is based on an original method proposed by Cundall and Strack [25]. The contact forces are calculated using Hertz-Mindlin no-slip contact model. It is based on a soft contact model or elastic approach, in which the magnitude of repulsive force is related of the amount of overlap. The normal force is calculated using a damped Hertzian normal contact model [26] with the damping term given by Tsuji et al. [27]. The magnitude  $F^n$  from a contact that resulted in a normal overlap  $\delta_n$  is given by:

$$F^n = -k_n \delta_n^{3/2} - \gamma_n \dot{\delta}_n \delta_n^{1/4}, \quad (2)$$

where  $k_n$  is the Hertzian normal stiffness coefficient,  $\delta_n$  is the deformation (normal particle overlap),  $\gamma_n$  is the normal damping coefficient, and  $\dot{\delta}_n$  is the rate of deformation.

In the above equation,  $k_n$  is obtained by:

$$k_n = \frac{4}{3} E_{eff} \sqrt{R_{eff}}, \quad (3)$$

where  $E_{eff}$  is the effective Young's modulus of two colliding entities (two particles or a particle and a wall). For entities with Poisson's ratios  $\nu_1$  and  $\nu_2$ , Young's moduli  $E_1$  and  $E_2$ ,  $E_{eff}$  is given by:

$$E_{eff} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2}, \quad (4)$$

where  $R_{eff}$  is defined as the effective radius of the contacting particles. In case of a particle-wall collision, the effective radius is simply the particle radius. While in the case of particle-particle collision, with the two contacting particles having radii  $R_1$  and  $R_2$ , the effective radius is obtained by:

$$R_{eff} = \frac{R_1 \times R_2}{R_1 + R_2}. \quad (5)$$

With the knowledge of the normal stiffness coefficient and a chosen coefficient of restitution  $\varepsilon$ , the normal damping coefficient  $\gamma_n$  is calculated as:

$$\gamma_n = 2 \sqrt{\frac{5}{3} \left[ \frac{\ln(\varepsilon) \times \sqrt{mk_n}}{\sqrt{\ln^2(\varepsilon) + \pi^2}} \right]}, \quad (6)$$

where  $k_n$  is the Hertzian normal stiffness coefficient.

Following the work of Mindlin and Deresiewicz [28], the tangential force  $F^t$  is calculated in a similar method as its normal counterpart. The tangential contact force also consists of elastic and damping components. When a tangential overlap of  $\delta_t$  is detected and there is a corresponding normal overlap of  $\delta_n$  due to the same contact, then the tangential force is expressed by:

$$F^t = -k_t \delta_t - \gamma_t \dot{\delta}_t \delta_n^{1/4}, \quad (7)$$

where  $k_t$  the tangential stiffness coefficient and  $\gamma_t$  is the tangential damping coefficient.

In the above equation,  $k_t$  is calculated by:

$$k_t = 8G_{eff} \sqrt{R_{eff} \sqrt{\delta_n}}, \quad (8)$$

where  $G_{eff}$  is the effective shear modulus. For two entities with shear moduli  $G_1$  and  $G_2$ ,  $G_{eff}$  is calculated as:

$$\frac{1}{G_{eff}} = \frac{2-\nu_1}{G_1} + \frac{2-\nu_2}{G_2}, \quad (9)$$

where  $\nu_1$  and  $\nu_2$  are the Poisson's ratios.

The tangential displacement (or overlap)  $\delta_t$  is calculated by time-integrating the relative velocity of tangential impact,  $v_{rel}^t$  between two colliding entities (either interparticle or particle-wall contact):

$$\vec{\delta}_t = \int v_{rel}^t dt. \quad (10)$$

The capabilities of EDEM include user defined functions and various features for simulating impregnation process, which has been developed in our previous work [17]. The fluid spray components are modeled as discrete droplets, which are sprayed from above the rotating bed and are absorbed upon contact with the simulated catalyst particles. The corresponding contact causes the simulated fluid droplet to essentially disappear while simultaneously transferring the mass of the fluid droplet to the simulated catalyst support particle, leading to a net increase in the mass. The mass flow rate of the fluid is defined as:

$$Q_{spr} = NV = N \frac{m}{\rho} \quad (11)$$

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