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Structural characterization of particle systems using spherical harmonics

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ABSTRACT

Many important properties of particulate materials are heavily influenced by the size and shape of the constituent particles. Thus, in order to control and improve product quality, it is important to develop a good understanding of the shape and size of the particles that make up a given particulate material. In this paper, we show how the spherical harmonics expansion can be used to approximate particles obtained from tomographic 3D images. This yields an analytic representation of the particles which can be used to calculate structural characteristics. We present an estimation method for the optimal length of expansion depending on individual particle shapes, based on statistical hypothesis testing. A suitable choice of this parameter leads to a smooth approximation that preserves the main shape features of the original particle. To show the wide applicability of this procedure, we use it to approximate particles obtained from two different tomographic 3D datasets of particulate materials. The first one describes an anode material from lithium-ion cells that consists of sphere-like particles with different sizes. The second dataset describes a powder of highly non-spherical titanium dioxide particles.

1. Introduction

Granular materials are used in many different industrial applications. For instance, they are used as ingredients in pharmaceutical materials and in the production of semiconductors and energy materials such as lithium-ion cells, solar cells and fuel cells [1–3]. Both the transport and industrial processing of agglomerates are greatly influenced by the shape and size of the agglomerate components [4]. For example, the flow, handling and rheology of granular materials are directly influenced by the shape and size of the constituent particles [1,5–7]. The performance of particulate materials also depends directly on their microstructure. For example, the performance of lithium-ion anodes depends strongly on the morphology of graphite particles and their spatial arrangement [8].

Tomographic three dimensional (3D) images are ideal sources for investigation of particle characteristics. Many different imaging techniques exist, including electron tomography [9,10] and focused ion-beam (FIB) tomography [11,12], which have resolutions on the nm-scale, synchrotron tomography [13] and X-ray microtomography (μ-CT) [14,15], which have resolutions on the µm-scale, and neutron-tomography [16] which can be used for investigations of larger objects.

Since particles in experimental 3D datasets are represented by sets of voxels, their analysis is a non-trivial task. In addition, particles can have rough surfaces and there are often artifacts present in the data, e.g., caused by the measurements or preliminary image processing steps like filtering and binarization. Thus, a different particle representation is needed to reproduce the properties of particle shapes, which is suitable for many materials. In some cases, this can done by using simple geometric objects like spheres, ellipsoids or unions of spheres. However, these simple objects cannot reproduce the shape of more complex particles, because important characteristics like volume, surface area or surface roughness are not preserved.

In this paper, we use the spherical harmonics expansion [17–19] to calculate an alternative representation of particles based on voxelized objects. Spherical harmonics have proven to be a valuable tool for the representation of particles [20,21]. The exact shape of a particle is represented as a combination of objects with growing roughness, the spherical harmonic functions. The spherical harmonic functions in the expansion are ordered in such a way that the roughness of the functions increases with the length of expansion. This kind of hierarchical representation is essentially influenced by a cutoff parameter to achieve a smooth approximation. The cutoff parameter is crucial as it controls the balance between the quality and the smoothness of the approximation. We present a method for optimally choosing this cutoff parameter, L, the length of expansion, based on statistical hypothesis testing. We show by comparing the mean square error that in this way an

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approximation is obtained which is in good accordance with the voxelized representation for complex shaped objects. Furthermore, we use an analytic description of this representation to calculate different particle characteristics and compare them to those obtained directly from the voxelized objects. A basic characteristic is the radius of an equivalent sphere, where this sphere can be defined to have equal volume, equal surface area or the same minimum or maximum particle axes, depending on the given application [22]. Other characteristics are sphericity [23] and characteristics that are based on the convex hull [24] and Gaussian curvature [25–27]. We note that the representation of particle shapes in spherical harmonics enables the definition and calculation of more refined characteristics as stated in [28]. Some of these characteristics can be linked with effective physical properties of the materials like diffusive behavior or interfacial reaction rates [8,29].

In order to demonstrate the potential and generality of this method we apply it to two different particle systems. Both samples are obtained using 3D imaging techniques with subsequent segmentation. The first particle system is extracted from the anode of a lithium-ion cell and consists of LiC_6 particles. The second sample describes a powder of highly non-spherical TiO_2 particles.

The rest of this paper is organized as follows. In Section 2, the class of spherical harmonic functions is introduced. We discuss the definition of the boundary for an object which is defined on a voxel grid and present an algorithm for its fast evaluation. For the purpose of implementation, all necessary algorithms and numerical details for the fast and efficient calculation of the coefficients in the spherical harmonics expansion are briefly recalled. Furthermore, we propose a method to estimate the parameter L, which determines the approximation quality and the smoothing effect in the expansion. In Section 3, this technique is applied to experimental data. After a short description of the materials, the approximation of particles from both samples by spherical harmonics is described. A comparison of the particle systems from the two different materials is performed using the spherical harmonics expansion. The goodness of approximation is discussed and various structural characteristics like particle sizes, surface areas and surface roughness are calculated. Finally, an outlook to further possibilities regarding the representation of particle systems by means of spherical harmonics concludes the paper.

2. Representation of particles by spherical harmonics

In this section we introduce the mathematical background of spherical harmonics and describe the techniques required for application to particles extracted from 3D images. Throughout this section, a particle is taken to be a set of connected voxels in a binary image, where each voxel can only adopt one out of two values which indicates whether the voxel belongs to the foreground or background, respectively. The two possible states are denoted by *true* and *false*. The spherical harmonics are a set of functions defined on the unit sphere which form a basis for a large class of functions. In fact, each square integrable function on the unit sphere can be represented as a series of spherical harmonics. In the situation where the functions define the boundary of the particles, this integrability condition is always naturally fulfilled. An important requirement for the particles is that they are star shaped (or star convex) with respect to a centroid [30], in our case to the barycenter. If this is true, it is possible to define a radius function on the unit sphere to fulfill the conditions for the expansion in spherical harmonic functions. The radius function maps each angle (θ, ϕ) on the unit sphere to the distance from the centroid to the boundary of the particle in that direction. Star shaped (or star convex) with respect to a point means that the connection from this point to each point of the particle lies completely inside the particle. This especially means that there are no holes or, e.g., curved intrusions into the particles. Furthermore, it only makes sense to calculate a smooth approximation of particles if it is reasonable to assume that the observed objects are smooth.

2.1. Definition and calculation of the boundary

During the preprocessing of data, the 3D images are binarized and segmented using a morphological segmentation method. In our case, a watershed transform [31–34] is used. This means that the binary image B is divided into distinct regions $B_1, \ldots, B_n$ with $\cup_{i=1}^{n} B_i = B$ and $B_i \cap B_j = \emptyset$ for $i \neq j$. The set of foreground voxels in a region corresponds to exactly one particle. In the following we describe the procedure that is applied to each particle.

The first step is to determine the distance from the barycenter of the particle to the boundary in each direction in order to compute the *radius function*. However, it is not clear how the boundary should be defined to model the original object as accurately as possible. There are several reasons for this. The discretization of the real object, based on grayscale intensities in an image, to Boolean values can be done using some kind of threshold to decide whether a voxel is classified as foreground or background. Thus, it is clear that the boundary cannot be defined without some assumptions about the preliminary step of discretization. Fig. 1 shows a 2D example of the consequences of different thresholds for the voxelized object. In the first case, shown in Fig. 1(a), every voxel that covers a part of the original object, which means that it has an intensity value larger than zero, is put to foreground which leads to an overestimation of the size of the object. In the other case, considered in Fig. 1(b), only voxels that are completely inside the object, which means that their intensity has the maximal value, are marked as foreground which leads to an underestimation of the size of the object.

Therefore, it is important to have information on the choice of the threshold and other preliminary steps for the binarization. In the extreme cases discussed above one can perform a morphological erosion or dilation [35] as a correction.

After a suitable preprocessing of the particle, we need to determine the exact distance from the centroid to the boundary for each direction on the unit sphere. As stated above, we assume that the particle is star shaped to ensure that the algorithm proposed below yields valid results in all cases. For a given angle, (θ, ϕ), we use nested intervals for an efficient evaluation of the particle boundary of the voxelized particles. We use the diagonal size, d, of the bounding box calculated for the original object as an upper bound for the radius in each direction. We then consider the following procedure:

(1) Construct a unit vector e in direction (θ, ϕ).
(2) Put the initial interval $[a, b] = [0, d]$.
(3) If $(a + b)/2 \cdot e$ belongs to the particle, then put $a = (a + b)/2$, otherwise set $b = (a + b)/2$.
(4) Repeat step 3 if $b - a > \tau$, where τ is some required (maximum) tolerance.
(5) The result is $r(\theta, \varphi) = (a + b)/2$.

A schematic illustration of this procedure is shown in Fig. 2. The advantage of nested intervals is that the runtime of the algorithm is, in practice, nearly independent of the particle size because the computational effort is logarithmic in the diameter of the particle's bounding box for a fixed tolerance. The boundary in direction (θ, ϕ) can then be represented in Cartesian coordinates relative to the centroid by

$$
\begin{aligned}
x &= r(\theta, \phi) \sin\theta \cos\phi, \\
y &= r(\theta, \phi) \sin\theta \sin\phi, \\
z &= r(\theta, \phi) \cos\theta.
\end{aligned}
\tag{2.1}
$$

2.2. Expansion in spherical harmonics

The set of spherical harmonic functions $\{Y_l^m : [0, \pi] \times [0, 2\pi) \to [0, \infty) : l, m \geq 0\}$ is a basis for the family of square integrable functions defined on the unit sphere. This means that the radius function for a given particle can be expanded in terms of spherical harmonics, if the particle is star