Estimation of specific surface area of particles based on size distribution curve

Yahya Ghasemi
PhD Student, Department of Civil, Environmental and Natural Resources Engineering, Luleå Technical University, Luleå, Sweden
(corresponding author: yahya.ghasemi@ltu.se)

Mats Emborg
Professor, Department of Civil, Environmental and Natural Resources Engineering, Luleå Technical University, Luleå, Sweden

Andrzej Cwirzen
Holder of Chair, Department of Civil, Environmental and Natural Resources Engineering, Luleå Technical University, Luleå, Sweden

Introduction
Concrete in the plastic state can be characterised by several parameters, among which workability is probably the most important one. This is influenced by the water requirement, which in turn is a function of the particle shape of aggregates and binders and their specific surface area. While it is known that the shape of fine particles has a significant effect on the water demand, there are uncertainties regarding how the various shape parameters would affect the specific surface area, mainly because up to now many of the shape parameters have not yet been clearly defined and there are no commonly accepted methods for their measurement and/or estimation. In this research, the actual particle shapes were replaced with regular convex polyhedrons to calculate the total specific surface area using the size distribution curves of the samples. The obtained results indicate that while, in some cases, the assumption of a spherical particle shape leads to an acceptable estimation of the specific surface area when compared with Blaine test results, the specific surface area of powders with more angular particles could be calculated more accurately with the assumption of a polyhedron shape rather than a sphere.

Notation

- $\omega_i$: mass of a grain fraction $i$, being the mass percentage of the fraction between $d_i$ and $d_{i+1}$
- $\rho_s$: specific density of the particles
- $d_{geo}$: geometric mean diameter of particles in fraction $i$
- $d_{arith}$: arithmetic mean diameter of particles in fraction $i$
- $d_i$: mean diameter of fraction $i$ and $i + 1$
- $SA_i/V_i$: specific surface area to volume ratio of fraction $i$
- $\alpha_{sph}$: calculated specific surface area of spherical particles
- $\alpha_{sph}$: calculated specific surface area of spherical particles
- $a_c$: circumsphered edge length
- $a_m$: midsphered edge length
- $d_{i, circ}:$ arithmetic mean diameter of particles in fraction $i$
- $d_{i, geo}:$ geometric mean diameter of particles in fraction $i$

Workability in the fresh state is one of the most important factors in design and production of concrete and can be related to the water demand of the mixture, which in addition to other factors is a function of the particle shape of aggregates and binders and their specific surface area. While it is known that the shape of fine particles has a significant effect on the water demand, there are uncertainties regarding how the various shape parameters would affect the specific surface area, mainly because up to now many of the shape parameters have not yet been clearly defined and there are no commonly accepted methods for their measurement and/or estimation. In this research, the actual particle shapes were replaced with regular convex polyhedrons to calculate the total specific surface area using the size distribution curves of the samples. The obtained results indicate that while, in some cases, the assumption of a spherical particle shape leads to an acceptable estimation of the specific surface area when compared with Blaine test results, the specific surface area of powders with more angular particles could be calculated more accurately with the assumption of a polyhedron shape rather than a sphere.

In asphalt mixtures, the SSA of the aggregate can be directly related to the asphalt concrete binder thickness, and is therefore related to the rutting and fatigue performance of asphalt concrete (Alexander and Mindess, 2010). Furthermore, Hunger (2010) concluded that, in the case of self-compacting concrete, a certain thickness of water layer surrounding the particles in the mixture will put the mixture at the onset of flow. In other words, the relative slump of a water–powder dispersion will put the mixture at the onset of flow. It is also possible to estimate the SSA using particle size distribution data based on the assumption that particles have spherical shapes. However, particle shapes are far from being spherical due to 3D randomness in their dimensions, related to the origin of the aggregates, and their production method. This is particularly true in the case of crushed aggregate.

The SSA is the quotient of the absolute available surface inclusive of all open inner surfaces (pore walls) divided by the
Increasing the fraction between di based on the particle size distribution and grading curves (McCabe et al., 1993),

\[ a_{sph} = 6 \sum_{i=1}^{n} \frac{o_i}{d_i \rho_s} \]

where \( o_i \) is the mass of a grain fraction \( i \), being the mass percentage of the fraction between \( d_i \) and \( d_{i+1} \); \( d_i \) is the mean diameter of fraction \( i \) and \( i+1 \); and \( \rho_s \) is the specific density of the particles.

Since the solid constituents of concrete mixtures seldom have a spherical particle shape, some error should be expected in the results from Equation 1. It has been found that the SSA of the aggregates can be much larger than that of spheres of equivalent size (Wang and Frost, 2003).

There are several ways of determining the SSA based on direct and indirect measurements – for example the Blaine test (ASTM C204 (ASTM, 2016)), and the Lea and Nurse method (Lea and Nurse, 1939). Both tests give similar results, but are not applicable to fine and ultra-fine powders. The Blaine test method was developed exclusively for measurement of the SSA of cement and is based on assuming a spherical particle shape, which leads to relative measures for materials other than cement.

Another method that has been used to determine the SSA is the volumetric static multi-point method, better known as the BET (Brunauer–Emmett–Teller) method (Brunauer et al., 1938). Results from the BET test include the measurement of the surface area of the internal pores, which is not of interest for calculation of water demand in concrete mixtures.

Determination of the SSA value using the aforementioned test methods includes complex measuring devices. As a result, developing a method that is cheaper and easier to use for estimation of the SSA is necessary. The main aim of this research was to verify the effect of the assumption of ideal polyhedron shapes for the particles, instead of spheres, on calculation of the SSA. For this purpose, the SSAs of the particles were mathematically calculated based on the size distribution curve and the assumption that particles have a uniform shape. The particle shapes were substituted with the shape of standard Platonic solids. The calculated values were compared to the SSA of the samples measured using the Blaine method.

### Table 1. Densities and SSA of the powders, from Hunger and Brouwers (2009) and Jennings et al., (1996)

<table>
<thead>
<tr>
<th>Material</th>
<th>Specific density: g/cm(^3)</th>
<th>Loose packing density</th>
<th>Bulk density: g/cm(^3)</th>
<th>SSA based on Blaine: cm(^2)/g</th>
</tr>
</thead>
<tbody>
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<td>CEM III/B 42.5 N</td>
<td>2.96</td>
<td>0.72</td>
<td>2.13</td>
<td>4500</td>
</tr>
<tr>
<td>Marble powder</td>
<td>2.80</td>
<td>0.64</td>
<td>1.79</td>
<td>4580</td>
</tr>
<tr>
<td>Limestone powder</td>
<td>2.21</td>
<td>0.69</td>
<td>1.87</td>
<td>4040</td>
</tr>
<tr>
<td>Quartz powder</td>
<td>2.60</td>
<td>0.64</td>
<td>1.66</td>
<td>2600</td>
</tr>
</tbody>
</table>

### Figure 1. Particle size distribution curves (Hunger and Brouwers, 2009; Jennings et al., 2013)

### Materials
Four types of powders were used in this study and all required input data were extracted from earlier test results. Characteristics of the first three materials shown in Table 1 were extracted from Hunger and Brouwers (2009) and included specific density, bulk density, grading curve, Blaine values, and scanning electron microscope (SEM) images. The information on the Quartz powder was obtained from Jennings et al. (2013).

The particle size distribution curve of the materials was obtained by deploying the low angle laser light scattering technique conducted by Hunger and Brouwers (2009), Figure 1.

Additionally, to further validate the relation between polyhedrons and the particle shape and to be able to distinguish the difference in particle geometry, SEM images from Hunger and Brouwers (2009) are shown in Figure 2.

### Computation of the SSA

#### Square-cube law
The square-cube law defines a mathematical principle describing the relationship between the volume and the area related to changes in size, and was first introduced by Galilei (Galilei...
and Drake, 1946). According to the principle, as a shape grows in size, its volume grows faster than its surface area. Consequently, as the size decreases, its surface area grows faster than its volume. The effect of the square-cube law becomes especially significant for calculation of the SSA of finer particles, namely powders and cement – that is, for a given mass of aggregate, the surface area increases with reducing particle size. The SSA can be calculated mathematically by the assumption of a spherical shape for the particle. In cases where spheres are replaced by another shape, the difference in calculations is caused by the fact that different shapes have different volumes, and also the ratio between SSA and volume changes based on the chosen shape according to the square-cube law. Figure 3 shows the difference in pace of growth of surface area/volume ratio (SA/V) of so called Platonic solids – a set of five 3D regular convex polyhedrons – obeying the square-cube law.

The formula presented in Equation 1 deals with a special case of calculating the SSA for spherical particles. The equation can be written in its general form, where the ratio of SA/V implements the square-cube law in the formula

\[
ad_{\text{poly}} = \sum_{i=1}^{N} \frac{SA_i \cdot \phi_i}{V_i \cdot \rho_s}
\]

where \( SA_i / V_i \) is the surface area to volume ratio of fraction \( i \) and is related to the shape as shown in Table 2.

As mentioned before, it is possible to calculate the SSA based on particle size distribution curves and with the assumption of mono-shaped particles. The Platonic solids that were examined to re-calculate the SSA are shown in Table 2. Substituting spheres with the Platonic solids will not only change the
calculated volume and SSA but will also affect the rate of growth in SA/V according to the square-cube law.

Equivalent polyhedron shape
The SSA of each fraction can be calculated using Equation 1. To do so, the mean diameter of the particle sizes $d_i$ and $d_{i+1}$ of a fraction $i$, as the characteristic particle size, is required. The mean diameter $d_{i,\text{arith}}$ can be calculated using either the arithmetic mean or geometric mean, as in Equations 3 and 4, respectively.

3. $d_{i,\text{arith}} = \frac{d_i + d_{i+1}}{2}$

4. $d_{i,\text{geo}} = \sqrt{d_i^2 + d_{i+1}^2}$

For the corresponding calculations of the polyhedrons, the length of the sides is needed since the Platonic solids should be defined in relation to the spheres. This relation can be conditioned based on the geometric properties of the spheres using the concepts of circumsphere and midsphere or by equivalent volume (mass) to the spheres. In geometry, a circumscribed sphere, or circumsphere, of a polyhedron is a sphere that contains the polyhedron and touches each of the polyhedron's vertices. A midsphere is defined as a sphere that touches all the polyhedron edges. The midsphere does not necessarily pass through the midpoints of the edges, but is rather only tangent to the edges at the same point along their lengths (Cundy and Rollett, 1981). The length of the edges of Platonic solids are smaller for the circumsphere approach compared to midsphere.

For volumetric equivalency, the sides of the polyhedrons can be back-calculated by replacing the volume of the polyhedrons by the volume of the spheres assumed for each fraction (Figure 4).

It is also possible to define the equivalency based on the concept of the insphere. The insphere is a sphere that is contained within the polyhedron and is tangent to each of the

Table 2. Platonic solids used in the calculation of SSA and their volumes

<table>
<thead>
<tr>
<th>Shape</th>
<th>Surface area</th>
<th>Volume</th>
<th>SA/V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tetrahedron</td>
<td>$\sqrt{3}a^2$</td>
<td>$\sqrt{2}a^3$</td>
<td>$\frac{14.697}{a}$</td>
</tr>
<tr>
<td>Cube</td>
<td>$6a^2$</td>
<td>$a^3$</td>
<td>$\frac{6}{a}$</td>
</tr>
<tr>
<td>Octahedron</td>
<td>$2\sqrt{3}a^2$</td>
<td>$\frac{1}{3}\sqrt{2}a^3$</td>
<td>$\frac{7.348}{a}$</td>
</tr>
<tr>
<td>Dodecahedron</td>
<td>$\sqrt{25 + 10\sqrt{5}}a^2$</td>
<td>$\frac{1}{4}(15 + 7\sqrt{5})a^3$</td>
<td>$\frac{2.694}{a}$</td>
</tr>
<tr>
<td>Icosahedron</td>
<td>$5\sqrt{3}a^2$</td>
<td>$\frac{5}{12}(3 + \sqrt{5})a^3$</td>
<td>$\frac{3.970}{a}$</td>
</tr>
<tr>
<td>Sphere</td>
<td>$4\pi a^2$</td>
<td>$4\pi a^3$</td>
<td>$\frac{3}{a}$</td>
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</table>

Figure 4. (a) Circumsphere, (b) midsphere and (c) volume equivalency of a cube
polyhedron’s faces. The issue with calculation based on the insphere is for some polyhedrons – for example the tetrahedron – only a relatively small sphere can be contained in comparison with other shapes. This would affect the calculation of the SSA, and therefore calculation based on the insphere was ignored.

In this study, different approaches have been examined to define the equivalent polyhedrons to the spheres by utilising the concepts of the circumsphere, midsphere, and volume equivalency. The computation was based on both arithmetic and geometric means.

The lengths of the sides of the polyhedrons were calculated for different assumptions.

- The polyhedrons are contained in spheres (circumsphere) with the diameter calculation based on the arithmetic mean.
- The polyhedrons are contained in spheres (circumsphere) with the diameter calculation based on the geometric mean.
- The sphere touches all the polyhedron edges (midsphere) with the diameter calculation based on the arithmetic mean.
- The sphere touches all the polyhedron edges (midsphere) with the diameter calculation based on the geometric mean.
- The polyhedrons have the same volume as the spheres (volumetric) with the diameter calculation based on the arithmetic mean.
- The polyhedrons have the same volume as the spheres (volumetric) with the diameter calculation based on the geometric mean.

The edge lengths of the polyhedrons, \( a \), were calculated by the equations listed in Table 3. The median radius of equivalent spheres, \( r \), can be calculated by either Equation 3 or 4.

### Results and discussion

Calculated SSAs according to Equation 2 and their corresponding Blaine values obtained from laboratory tests of the studies referred to previously are presented in Table 4 where the SSA was calculated based on the size distribution curve assuming different Platonic solids.

In the case of most of the studied powders, the calculated spherical values of SSA were close to the Blaine value. The marble powder showed the largest deviation, and thus its calculation should be conducted with the assumption of a different shape other than spherical. This result corresponds well to the observed elongated, flaky particle shape of marble powder (Figure 2).

Moreover, in the cases of CEM III/B 42·5 N, limestone powder and quartz powder, the calculation based on the assumption of a spherical shape for the particles led to an overestimation of SSA. It should be noted that, for the same volume, the less spherical a particle is, the greater is its SSA. Since the particle shapes of powders are normally anything but spherical, the SSA of the actual particles should therefore be higher than that calculated based on the assumption of spherical shape. A slight overestimation of spherical SSA can be related to the approach that is taken in determining the mean diameter of a particle.

In the case of marble powder, the calculated spherical SSA differs most noticeably from the Blaine value and is also the least spherical in terms of particle shape (Figure 2). It should also be mentioned that the Blaine test is a relative test designed for measuring the SSA of cement and not necessarily any non-spherical powder; in other words, the Blaine value is a relative value and not an absolute one.

To sum up, among the studied scenarios used for defining the equivalent shape and mean diameter, the assumption of midsphere equivalency and arithmetic mean results in less...
Table 4. Calculated SSA of the powders

<table>
<thead>
<tr>
<th>Material</th>
<th>Blaine Calculation based on circumsphere and arithmetic mean</th>
<th>Blaine Calculation based on circumsphere and geometric mean</th>
<th>Blaine Calculation based on midsphere and arithmetic mean</th>
<th>Blaine Calculation based on midsphere and geometric mean</th>
<th>Blaine Calculation based on equivalent volume and arithmetic mean</th>
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</table>
 error compared to other approaches (see the compilation in Figure 5).

As can be seen in Figure 5, the assumption of spherical shape agrees with the Blaine values for CEM III and quartz. While cement particles are usually round, the same cannot be said for quartz. The reason that the SSA calculated for quartz agrees with the spherical shape could be related to the fact that the source of information for quartz comes from different research (Jennings et al., 2013).

In the case of marble powder, an assumption of cubical shape leads to a better estimation of SSA, which can be directly related to the angularity of marble grains. The calculations based on a cubical shape rather than spherical for marble powder shows a 40% increase in SSA; in that sense, assuming two identical concrete recipes at a water:cement ratio of 0.5, with the only difference being the assumption made for the general shape of particles for the marble powder, the concrete made with cubical particles requires around 5 more litres of water in a cubic metre of concrete for the mixture to be put at the onset of flow (according to the excess water layer theory, with a water layer thickness of 25 nm, as mentioned by Hunger and Brouwers (2009)) compared to the water requirement of spherical particles, which illustrates the significance of replacing the spherical shape with a more representative shape. Moreover, it should be noted that for the finer particles, there is a larger difference in the calculated SSA for different shapes, which can be related to the principle of the square-cube law.

It should also be mentioned that each column of data in Figure 5 shows the difference in calculated SSA based on a different shape for a given size distribution curve. The difference in SSA for different shapes becomes more significant as the fine content of the studied materials increases as a result of the square-cube law; for example, see the difference in SSA for limestone compared to quartz.

**Conclusion**

The SSA of aggregates, fillers and binders affects the fresh and hardened concrete properties. The water layer theory is a potentially useful tool for prediction of fresh concrete flowability. However, the complexity of the analytical instruments required to measure the SSA limits wider use of that approach for example, in concrete mix design. An alternative is to formulate an equation enabling theoretical prediction of the SSA using only simple input data. The results of the present study showed that while the assumption of spherical shape for particles leads to an acceptable estimation of the SSA for round particles, in the case of more angular, flaky particles, substituting polyhedrons for spheres improves the accuracy of SSA estimation. The significance of the study lies in the fact that in almost all approaches taken for quantifying the shape, such as sphericity (the ratio of the SA of a material to the SA of a sphere of the same volume), the concept of the square-cube law and changes in the pace of growth of the SA to volume ratio is overlooked, while in the proposed approach of replacing spheres with standard 3D shapes, the size of fractions and the square-cube law are implemented.

The concept described in this paper is being used to develop a mix design model based on the size distribution curve, packing density and water layer theory principles. Some pilot concrete tests conducted in the laboratory, based on the model, showed promising results.

**Acknowledgement**

The authors would like to thank Dr Martin Hunger for his guidance and the Swedish Research Council (Formas) for the financial support.

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