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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

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.

Notation

a_c	circumsphered edge lentgh
a_m	midsphered edge lentgh
$d_{i,arith}$	arithmetic mean diameter of particles in fraction i
$d_{i,geo}$	geometric mean diameter of particles in fraction i
$\bar{d}_i$	mean diameter of fraction i and $i+1$.
SA_i/V_i	specific surface area to volume ratio of fraction i
α_{sph}	calculated specific surface area of spherical particles
ρ_s	specific density of the particles.
ω_i	mass of a grain fraction i, being the mass percentage of the fraction between d_i and d_{i+1}

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 aggregates' shape, size, and fines content. Thus, understanding the role of aggregates is fundamental to the production of high-performance concrete (Alexander and Mindess, 2010).

Aggregates have a large variability in mineral composition, shape, surface roughness and texture, and specific surface area (SSA). One major parameter influencing water demand is a comprehensive measurement of size, shape, and roughness (Wang and Lai, 1998).

The shape of particles is a complex function of their formation conditions, the mineralogical composition and particle size, and not only refers to the basic shape of the aggregates, but also to other characteristics such as angularity, flakiness, and so on. There is a considerable confusion about how various shape parameters are defined. There are also no commonly accepted methods for their measurement (Kwan and Mora, 2002). Particle shape can be classified by measuring the particles' length, width, and thickness; estimation is easier for larger particles. The SSA can be used as an indicator of their size, shape, and surface roughness.

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 water–powder dispersion will put the mixture at the onset of flow. In other words, the relative slump of a water–powder mixture becomes a function of the SSA when sufficient water is present to enable the flow (Brouwers and Radix, 2005).

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 mass (m^2/g). For concrete mix design, only the outer surface in contact with water is of interest. With the consideration of the specific density, the SSA could also be expressed as area per volume (m^2/m^3). The total surface area of a set of aggregates is governed by the finer fractions of aggregates and powders according to the square-cube law. Assuming all particles were spherical in shape, the SSA, α_{sph}, would be easy to calculate based on the particle size distribution and grading curves (McCabe *et al.*, 1993),

$$1. \quad a_{\mathrm{sph}} = 6\sum_{i=1}^{n}\frac{\omega_i}{\bar{d}_i \cdot \rho_{\mathrm{s}}}$$

where ω_i is the mass of a grain fraction i, being the mass percentage of the fraction between d_i and d_{i+1}; $\bar{d}_i$ is the mean diameter of fraction i and $i+1$; and ρ_{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)

Material	Specific density: g/cm^3	Loose packing density	Bulk density: g/cm^3	SSA based on Blaine: cm^2/g
CEM III/B 42·5 N	2·96	0·72	2·13	4500
Marble powder	2·80	0·64	1·79	4580
Limestone	2·21	0·69	1·87	4040
Quartz powder	2·60	0·64	1·66	2600

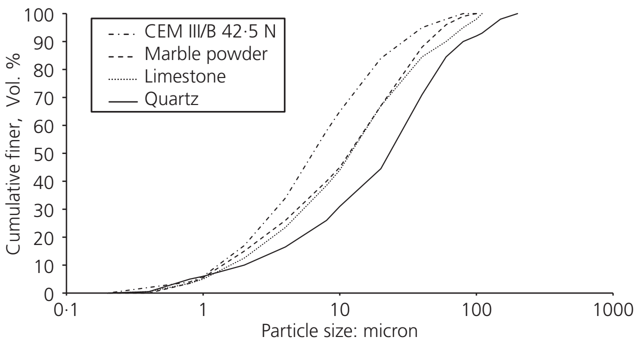


Figure 1. Particle size distribution curves (Hunger and Brouwers, 2009; Jennings *et al.*, 2013)

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

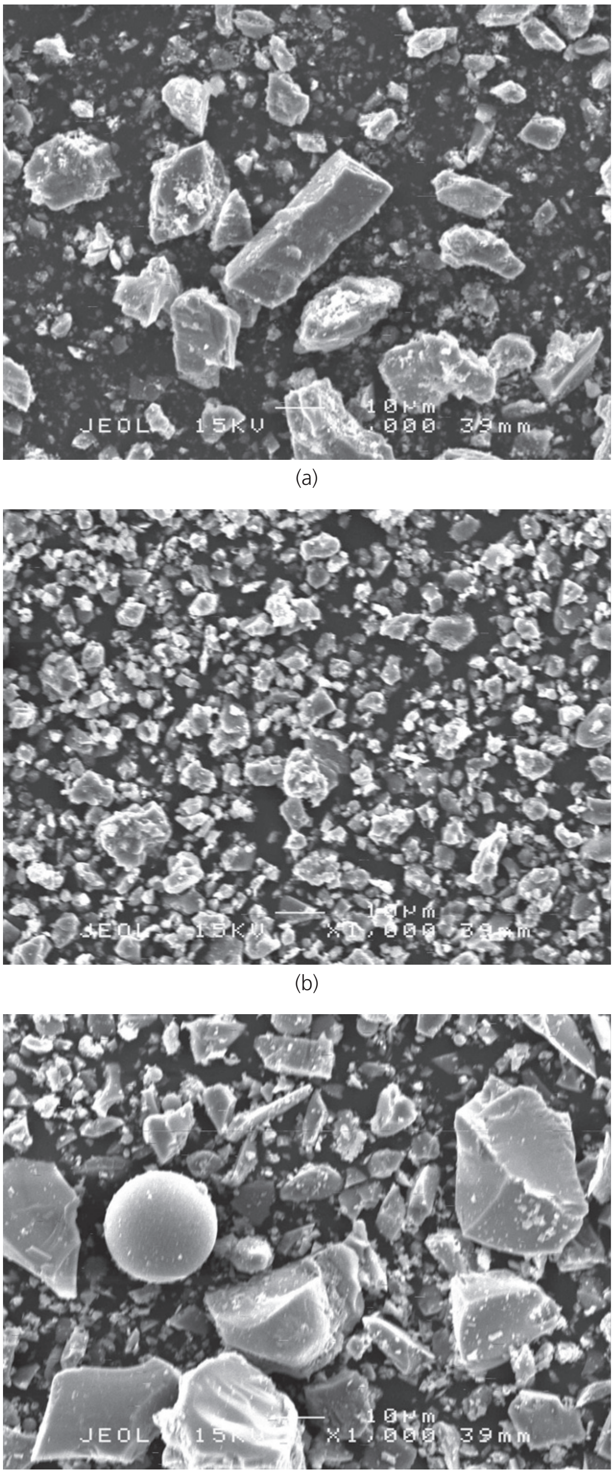


Figure 2. SEM images of studied powders, 1000 × magnification. (a) Dolomite marble powder, (b) limestone powder, (c) CEM III/B 42·5 N (Hunger and Brouwers, 2009)

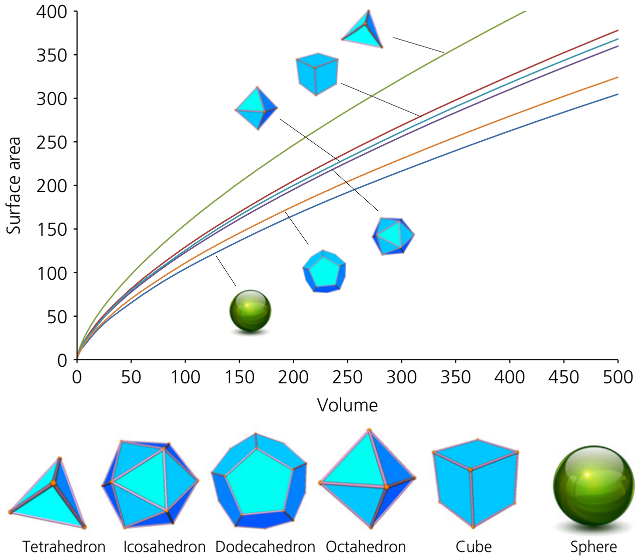


Figure 3. Surface area plotted against volume of the Platonic solids and a sphere (Ghasemi *et al*., 2016)

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.

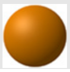
The formula presented in Equation 1 deals with a special case of calculating the SSA for spherical particles. The equation can be written in its general form, where the ratio of SA/V implements the square-cube law in the formula

2. $$a_{\text{poly}} = \sum_{i=1}^{n} \frac{\text{SA}_i.\omega_i}{\text{V}_i.\rho_\text{s}}$$

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

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

Shape		Surface area	Volume	SA/V
Tetrahedron		$\sqrt{3}a^2$	$\frac{\sqrt{2}a^3}{12}$	$\frac{14{\cdot}697}{a}$
Cube		$6a^2$	a^3	$\frac{6}{a}$
Octahedron		$2\sqrt{3}a^2$	$\frac{1}{3}\sqrt{2}a^3$	$\frac{7{\cdot}348}{a}$
Dodecahedron		$\sqrt[3]{25+10\sqrt{5}}a^2$	$\frac{1}{4}(15+7\sqrt{5})a^3$	$\frac{2{\cdot}694}{a}$
Icosahedron		$5\sqrt{3}a^2$	$\frac{5}{12}(3+\sqrt{5})a^3$	$\frac{3{\cdot}970}{a}$
Sphere		$4\pi a^2$	$\frac{4\pi a^3}{3}$	$\frac{3}{a}$

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 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

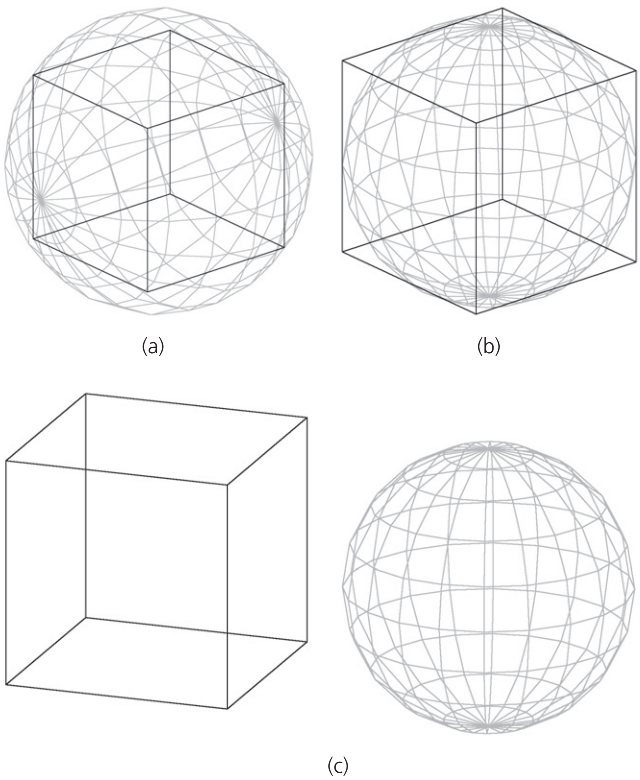


Figure 4. (a) Circumsphere, (b) midsphere and (c) volume equivalency of a cube

Table 3. Edge lengths of polyhedron

Shape		Circumsphered edge length: a_c	Midsphered edge length: a_m
Tetrahedron		$\frac{4r}{\sqrt{6}}$	$\frac{4r}{\sqrt{2}}$
Cube		$\frac{2r}{\sqrt{3}}$	$\frac{2r}{\sqrt{2}}$
Octahedron		$\frac{2r}{\sqrt{2}}$	$2r$
Dodecahedron		$\frac{4r}{\sqrt{3}(1+\sqrt{5})}$	$\frac{4r}{(3+\sqrt{5})}$
Icosahedron		$\frac{4r}{\sqrt{10+2\times\sqrt{5}}}$	$\frac{4r}{(1+\sqrt{5})}$

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.

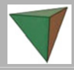
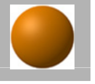
In the case of most of the studied powders, the calculated spherical values of SSA were close to the Blaine value. The marble powder showed the largest deviation, and thus its calculation should be conducted with the assumption of a different shape other than spherical. This result corresponds well to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

Material	Blaine	SSA: cm²/g					
							
Calculation based on circumsphere and arithmetic mean							
CEM III/B 42·5 N	4500	13 874	8014	8014	5822	5833	4625
Marble	4580	9596	5542	5543	4027	4036	3199
Limestone	4040	15 478	8940	8940	6495	6512	5160
Quartz	2600	8116	4689	4688	3397	3415	2705
Calculation based on circumsphere and geometric mean							
CEM III/B 42·5 N	4500	14 642	8458	8454	6140	6143	4881
Marble	4580	10 112	5838	5838	4240	4245	3371
Limestone	4040	16 325	9426	9426	6846	6851	5441
Quartz	2600	8562	4944	4944	3581	3594	2854
Calculation based on midsphere and arithmetic mean							
CEM III/B 42·5 N	4500	8011	6543	5664	5433	5215	4625
Marble	4580	5540	4525	3917	3761	3644	3199
Limestone	4040	8936	7300	6319	6064	5812	5160
Quartz	2600	4685	3828	3313	3005	2995	2705
Calculation based on midsphere and geometric mean							
CEM III/B 42·5 N	4500	7625	6904	5978	5743	5224	4881
Marble	4580	5164	4767	4128	3969	3608	3371
Limestone	4040	8534	7696	6665	6404	5824	5442
Quartz	2600	4656	4037	3495	3354	3054	2584
Calculation based on equivalent volume and arithmetic mean							
CEM III/B 42·5 N	4500	6892	5737	5469	5078	4927	4625
Marble	4580	4766	3968	3784	3515	3406	3199
Limestone	4040	7689	6401	6103	5668	5495	5160
Quartz	2600	4031	3355	3200	2964	2881	2705
Calculation based on equivalent volume and geometric mean							
CEM III/B 42·5 N	4500	7273	6056	5772	5358	5195	4881
Marble	4580	5022	4182	3986	3703	3588	3371
Limestone	4040	8109	6751	6434	5977	5791	5442
Quartz	2600	4253	3541	3375	3128	3037	2584

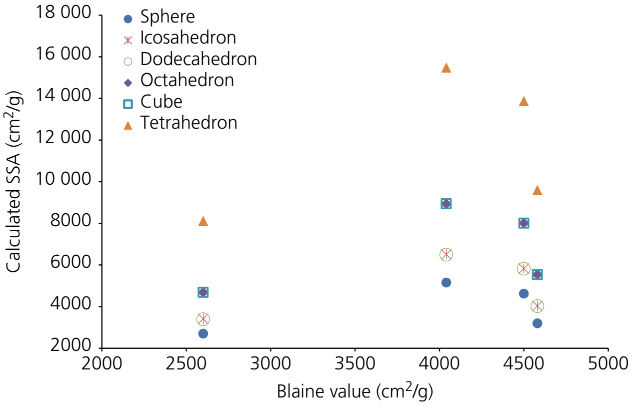


Figure 5. Blaine values plotted against calculated SSA based on midsphere-arithmetic mean assumption

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.

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