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Should Melamed's spherical model of size–colour dependence in powders be adapted to non-spheric particles?

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Abstract

In 1963, Melamed proposed a model that expressed reflectance of a powder described as a population of spherical particles of unique diameter as a function of size, shape and optical characteristics of the powder. This paper shows how, assuming particles to be ellipsoids of revolution, one dimension can be added. We show that the mean value of the shape coefficient of the model tends to converge if many particles are accounted for.

Keywords: Reflectance; Powders; Shape; Melamed; Particles arrangement

1. Introduction

Reflectance of powders is something hard to control as it depends on many parameters. These parameters are both intrinsic and extrinsic to the powder. It is well known that reflectance of a powder will depend on the size of the particles, on their shape and on the nature of the material. It will also depend on the way that the powder is prepared for the measurement, i.e. its compacity and the state of the surface. The reflectance spectrum is characteristic (under the geometrical conditions of the measurement) of the sample as it expresses the percentage of energy reflected by the sample related to the energy received by the sample from the lighting source. Reflectance does not take into account the characteristics of the human perception so it is not directly a measure of the perceived colour of a material but it is rather simple, knowing the reflectance to get the $L^*a^*b^*$ coordinates, to characterise the colour of the materials. The interested reader should refer to [1,2] for more details.

In 1963, Melamed [3] developed a model that directly expresses reflectance as a function of different parameters such as particle diameter d , refractive index n , absorption coefficient

k , wavelength of the incident radiation λ , and a coefficient (x_u) that depends on particle shape and arrangement.

This model is very different from the well-known Kubelka–Munk model [4] which is commonly used in industry for colour-matching problems. Kubelka–Munk's model is a “continuous” model, i.e. it considers that the medium, even if composed of different components, is one material with its own optical properties. Its main advantage is to allow indirect reflectance summation because the coefficient of diffusion S and of absorption K are additive and are related to the reflectance through the Kubelka–Munk function.

Melamed's model is a “discontinuous” model as it considers that the material is composed of particles with their own physical and optical properties. The reflectance is then an individual property of the particle, and the aim is to make it become a collective property of the material by taking into account the arrangement of the particles, through the coefficient x_u . Melamed calculated x_u value for spheres of unique diameter and arranged in a compact hexagonal way.

Since this ideal case never applies to real, industrial powders, we want to enlarge the use of this model by adapting [5] it to populations of particles of any shape, with a granulometric distribution, and randomly arranged. The coefficient x_u can be supposed to be dramatically depending

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on particle shape and that is what we want to verify within the scope of this study. Other fits of the model are presented in [6].

As will be described, the method used is both mathematical and experimental: mathematical because some calculations are necessary and experimental because, knowing the future use of the coefficient, some simplifications have been made possible.

2. Definition of x_u [7]

x_u is a geometrical parameter depending on particle shape and arrangement. It represents the probability that a ray coming from the centre of one particle emerges upwards without being reflected by the surface of a neighbouring particle.

We consider that a real sample is made of numerous particles arranged randomly so that, theoretically, x_u is different for each particle. For one central particle surrounded by its close neighbours, the problem is to calculate the part of the open space that is occupied by these neighbours which therefore prevent rays coming from the centre of the central particle from emerging upwards. This part of occupied space is linked to the notion of solid angle. Fig. 1 defines the angle α that will serve in the calculation of the solid angle and shows that this angle strongly depends on the shape of particles.

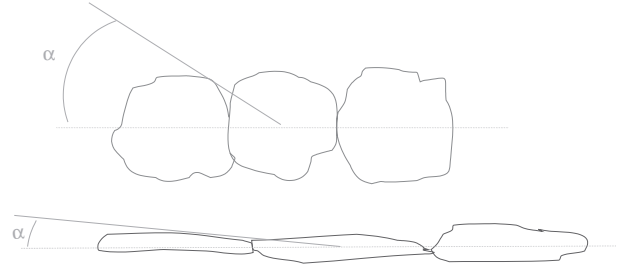


Fig. 1. Angle α for two different shapes of particles.

In the literature, the case of arrangement of spheres of uniform size (or not) has often been treated [8]. We can find many examples of modelling of arrangements of such particles. As real particles are sometimes far from this shape, we wanted to enlarge the study to be able to apply the model for other types of particles. We chose ellipsoids of revolution, which allow us to add one dimension to the problem. In the literature, we can find some examples of arrangement of ellipsoids [9–11], but these arrangement are not randomly constructed, they are composed of a repeating geometric pattern.

3. General approach

Fig. 2 schematically shows our general approach for the calculation of the coefficient x_u .

4. Calculations details

As x_u is related to the solid angle, this notion should be developed:

4.1. Definition of the solid angle

Solid angle is defined as the three-dimensional ‘angle’ formed by the vertex of a cone (Fig. 3). For the case presented in Fig. 3, the calculation is rather simple. The solid angle ω is expressed as a function of the half-angle of the cone γ .

$$\omega = 2\pi(1 - \cos\gamma) \quad (1)$$

with

$$\tan\gamma = \frac{r}{OO'} \quad (2)$$

4.2. Calculation of x_u for spheres

This calculation for spheres is the simplest case because of symmetry. Fig. 4 shows a top view of a central sphere surrounded by its six neighbours. Fig. 5 shows a side view of a section of the same configuration.

As x_u represents the geometrical probability for upward diffusion, it can be expressed as the remaining space after the space occupied by the six neighbours of the central particle has been removed from the 2π steradians of the upper half space. The filled space corresponds for each particle to half of the solid angle ω . Thus x_u can be expressed as:

$$x_u = \frac{(2\pi - 6 \cdot \frac{\omega}{2})}{4\pi} \quad (3)$$

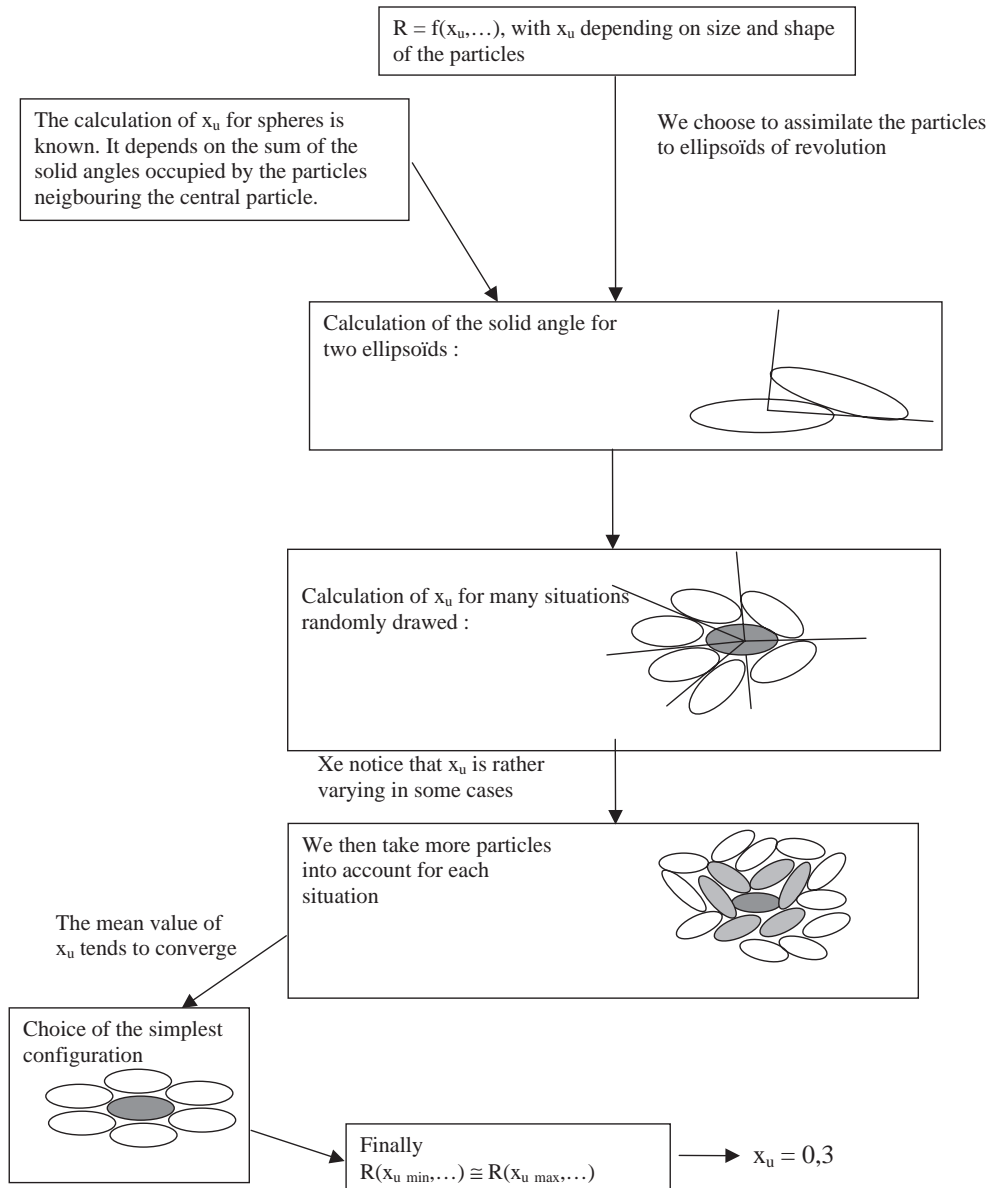


Fig. 2. General approach for the calculation of x_u .

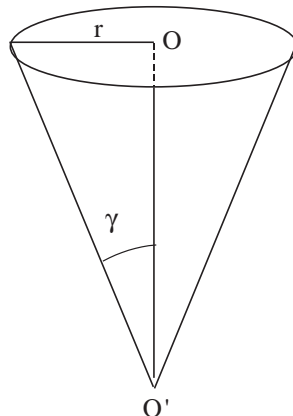


Fig. 3. Notion of solid angle.

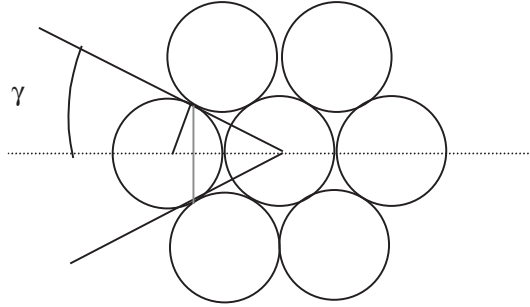


Fig. 4. Top view of one layer of hexagonal compact arranged spheres.

Knowing that for spheres γ is equal to $\pi/6$, we get for x_u :

$$x_u = 0.299.$$

This rather simple calculation can be applied only when the particles are spherical, because the surface intercepted by the two tangents issued from the centre of the sphere is then a disk, identical for each surrounding particle.

4.3. Calculation for ellipsoids of revolution

The case of ellipsoids is more complicated because each neighbouring ellipsoid occupies, when “viewed from” the central particle, a different space. As we suppose all the ellipsoids to be the same size, a top view shows ellipses in contact. All these ellipses are contained in the plane containing all their centres (Fig. 6).

In this case, the surface intercepted by the two tangents issued from the centre of the central ellipsoid is not a disk and the cone is not a cone of revolution.

To estimate x_u for this type of configuration, we nevertheless calculate the surface intercepted by the tangents (Fig. 7). This surface corresponds to the cross section of the ellipsoid containing (CC'). We then calculate the diameter of the disk whose surface is equal to the surface of this cross section and so we find again the same theoretical case as the one before.

4.3.1. Calculation of the surface of the cross section and of the solid angle

First, we consider two ellipses: one (denoted E') supposed to be the central one, and one of its neighbours, denoted (E). Their position is randomly defined. We define on each neighbour two orthonormal axes: the x -axis corresponding to the major axis of the ellipse and the y -axis to the minor axis. The centre is the centre of the ellipse. Thus, the centre O' of the central ellipse is defined by its co-ordinates $(x_{O'}, y_{O'})$ in this system of co-ordinates that we are going to use to define some geometrical objects useful in obtaining the solution.

The equation of the straight line (OO') is

$$x_{O'}y - y_{O'}x = 0. \quad (4)$$

The equation of the ellipse (E) is

$$\frac{x^2}{a^2} + \frac{y^2}{b^2} - \frac{1}{4} = 0 \quad (5)$$

with a and b respectively the major and minor axes of the ellipse.

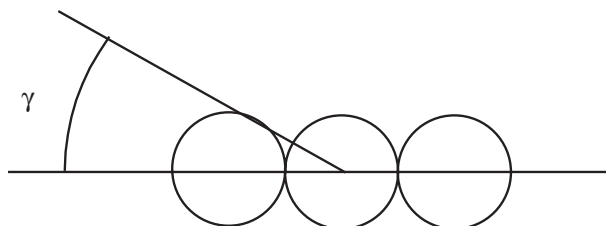


Fig. 5. Side view from one sphere and the space occupied by its neighbours (section).

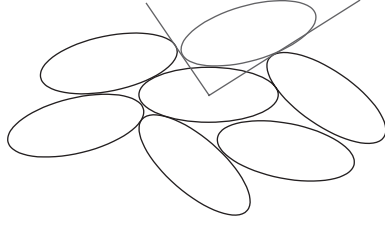


Fig. 6. Top view of an arrangement of ellipsoids (one layer).

The general equation of the tangent to a plane curve of equation $f(x,y)=0$ at a point (x',y') can be written:

$$\frac{\partial f}{\partial x}(x',y')(x-x') + \frac{\partial f}{\partial y}(x',y')(y-y') = 0 \quad (6)$$

and so the equation of the tangent line to (E) at the point M (x_M, y_M) is:

$$\frac{x_M}{a^2}(x-x_M) + \frac{y_M}{b^2}(y-y_M) = 0. \quad (7)$$

Two tangent lines at (E) containing O' can be drawn. If B and C are the two tangential points, the equations of the tangent lines are:

$$\left(\frac{x_{B,C}}{a^2}x + \frac{y_{B,C}}{b^2}y - \frac{x_{B,C}^2}{a^2} + \frac{y_{B,C}^2}{b^2} \right) = 0. \quad (8)$$

According to their definition, B and C belong to the ellipse, so the following equation is verified:

$$\frac{x_{B,C}^2}{a^2} + \frac{y_{B,C}^2}{b^2} = \frac{1}{4} \quad (9)$$

$$x_{B,C} = \pm a \sqrt{\frac{1}{4} - \frac{y_{B,C}^2}{b^2}}. \quad (10)$$

O' belongs to the two tangent lines so:

$$\frac{\pm a \sqrt{\frac{1}{4} - \frac{y_{B,C}^2}{b^2}}}{a^2} x_{O'} + \frac{y_{B,C}}{b^2} y_{O'} - \frac{1}{4} = 0 \quad (11)$$

and therefore,

$$\frac{\pm \sqrt{\frac{1}{4} - \frac{y_{B,C}^2}{b^2}}}{a} x_{O'} = \frac{1}{4} - \frac{y_{B,C}}{b^2} y_{O'} \quad (12)$$

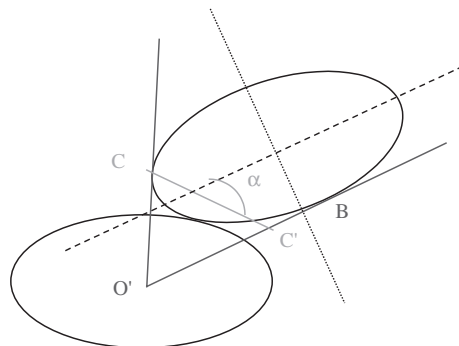


Fig. 7. Definition of the cross section of the cone.

or, squaring this equation,

$$y_{B,C}^2 \left(\frac{y_{O'}^2}{b^4} + \frac{x_{O'}^2}{a^2 b^2} \right) - y_{B,C} \left(\frac{y_{O'}}{2b^2} \right) + \frac{1}{16} - \frac{1}{4} \frac{x_{O'}^2}{a^2} = 0 \quad (13)$$

y_B and y_C are the two solutions of this quadratic equation. Arbitrarily, we choose $y_C < y_B$. We can calculate:

$$x_{B,C} = \pm a \sqrt{\frac{1}{4} - \frac{y_{B,C}^2}{b^2}}. \quad (14)$$

The sign of $x_{B,C}$ depends on the position of O' relative to the ellipse E. Three cases can be differentiated (cf. Fig. 8).

To estimate the solid angle, we have seen that we have calculated the surface of the vertical section of the cone containing (CC') . C' is the point belonging to $(O'B)$ and characterised by $O'C = O'C'$.

Let us find the co-ordinates of C' :

$$\begin{cases} C'(x_{C'}, y_{C'}) \in (O'B) \Rightarrow (y_B - y_{O'})x_{C'} - (x_B - x_{O'})y_{C'} - x_{O'}y_B + x_B y_{O'} = 0 \\ d(O', C') = d(O', C) \Rightarrow (x_C - x_{O'})^2 + (y_C - y_{O'})^2 = (x_{C'} - x_{O'})^2 + (y_{C'} - y_{O'})^2 \end{cases} \quad (15)$$

If $y_B = y_{O'}$ then, $y_{C'} = y_{O'}$, and we have a quadratic equation where $x_{C'}$ is the unknown quantity.

Otherwise, we have a quadratic equation where $y_{C'}$ is the unknown:

$$\begin{aligned} & \left(\frac{x_B - x_{O'}}{y_B - y_{O'}} \right)^2 + 1 \Big) y_{C'}^2 + 2 \left(\frac{x_B - x_{O'}}{y_B - y_{O'}} \right) \left(\frac{x_{O'} y_B - y_{O'} x_B}{y_B - y_{O'}} - x_{O'} \right) y_{C'} + \left(\frac{x_{O'} y_B - y_{O'} x_B}{y_B - y_{O'}} - x_{O'} \right)^2 \\ & + y_{O'}^2 - (x_C - x_{O'})^2 - (y_C - y_{O'})^2 = 0 \end{aligned} \quad (16)$$

Let H be the middle of $[CC']$. According to these definitions, we have $(O'H) \perp (CC')$.

The required surface is the intersection of the cone whose vertex is O' with the vertical plane containing (CC') .

A cone is a set of lines, all of them passing through a common point, called the vertex of the cone. These lines are called the generating lines. We can determinate an equation of the cone, knowing the co-ordinates of the vertex and the equation of one directrix of the cone. A directrix is a curve that cuts all the generating lines of the cone. If A (a, b, c) is the vertex of the cone and if one of its directrix can be defined by

$$\begin{cases} f(x, y, z) = 0 \\ g(x, y, z) = 0 \end{cases} \quad (17)$$

then an equation of the cone is:

$$\begin{cases} f(a + \lambda(x - a), b + \lambda(y - b), c + \lambda(z - c)) = 0 \\ g(a + \lambda(x - a), b + \lambda(y - b), c + \lambda(z - c)) = 0 \end{cases} \quad (18)$$

In the present case, the cone is of vertex O' and its generating lines are all the tangent lines to the ellipsoid passing through O' . One directrix of the cone is the intersection of the ellipsoid and the vertical plane containing the straight line (BC) .

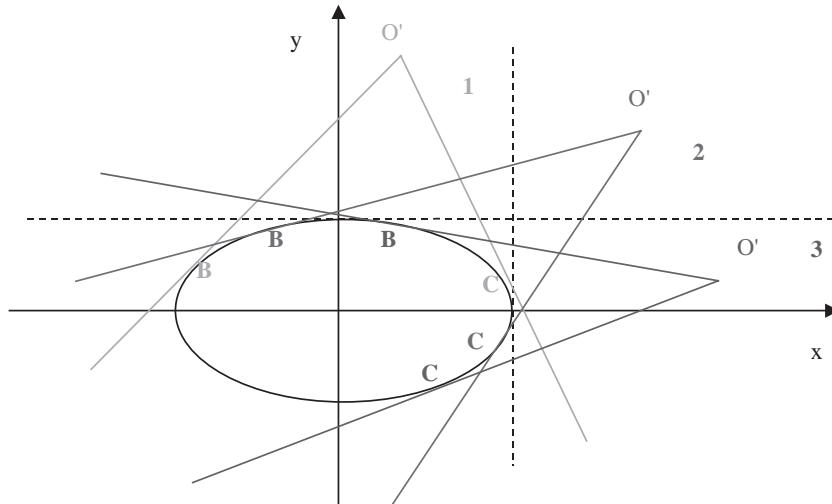


Fig. 8. Delimitation of the plane into three zones according to the position of O' relative to the ellipse.

Let us find the equation of this directrix:

- Equation of the vertical plane containing (BC)): one point M (x,y,z) belongs to this plane if:

$$\begin{vmatrix} 0 & x_B - x_C & x - x_B \\ 0 & y_B - y_C & y - y_B \\ 1 & 0 & z \end{vmatrix} = 0 \quad (19)$$

so:

$$(y_B - y_C)x - (x_B - x_C)y + x_B y_C - y_B x_C = 0 \quad (20)$$

- Intersection of this plane and of the ellipsoid of revolution:

$$\begin{cases} (y_B - y_C)x - (x_B - x_C)y + x_B y_C - y_B x_C = 0 \\ \frac{x^2}{a^2} + \frac{y^2}{b^2} + \frac{z^2}{b^2} - \frac{1}{4} = 0 \end{cases} \quad (21)$$

We define:

$$A1 = y_B - y_C$$

$$B1 = x_C - x_B$$

$$C1 = y_B x_C - x_B y_C \quad (22)$$

We then get a curve define by:

$$\begin{cases} y = \frac{C1}{B1} - \frac{A1}{B1}x \\ z^2 = -x^2 \left(\frac{b^2}{a^2} + \frac{A1^2}{B1^2} \right) + \frac{2A1C1}{B1^2}x + \frac{b^2}{4} - \frac{C1^2}{B1^2} \end{cases} \quad (23)$$

The vertex of the cone is $O'(x_{O'}, y_{O'}, 0)$, and one directrix is the curve defined by the equation before. So we can write: $M(x,y,z) \in (C)$ if and only if $\exists \lambda$ so that

$$\begin{cases} \frac{A1}{B1}(x_{O'} + \lambda(x - x_{O'})) + (y_{O'} + \lambda(y - y_{O'})) - \frac{C1}{B1} = 0 \quad (a) \\ (x_{O'} + \lambda(x - x_{O'}))^2 \left(\frac{b^2}{a^2} + \frac{A1^2}{B1^2} \right) - 2 \frac{A1C1}{B1^2}(x_{O'} + \lambda(x - x_{O'})) + \lambda^2 z^2 - \frac{b^2}{4} + \frac{C1^2}{B1^2} = 0 \quad (b) \end{cases} \quad (24)$$

An equation of the vertical plane passing containing the straight line (CC') is:

$$(y_C - y_C)x - (x_C - x_C)y + y_C x_C - x_C y_C = 0 \quad (25)$$

To find the intersection, we shall eliminate λ in Eq. (24b). And first, we shall express λ as a function of x and y using Eq. (24a), and then y as a function of x using Eq. (24b). This leads to:

$$\begin{cases} \lambda = \frac{\frac{C1}{B1} - \frac{A1}{B1}x_{O'} - y_{O'}}{(x - x_{O'}) \frac{A1}{B1} + \frac{y_C - y_{C'}}{x_C - x_{C'}}x + \frac{y_{C'}x_C - y_C x_{C'}}{x_C - x_{C'}} - y_{O'}} \\ z^2 = \frac{1}{\lambda^2} \left[-\frac{C1^2}{B1^2} + \frac{b^2}{4} + 2 \frac{A1C1}{B1^2}(x_{O'} + \lambda(x - x_{O'})) - (x_{O'} + \lambda(x - x_{O'}))^2 \left(\frac{b^2}{a^2} + \frac{A1^2}{B1^2} \right) \right] \end{cases} \quad (26)$$

This curve belongs to the vertical plane containing (CC'). In this plane:

$$S = \int_{x'_{C'}}^{x'_{C'}} \int_{-z'(x)}^{z'(x)} dx' dz' \quad (27)$$

so, considering the symmetry

$$S = \int_{x'_{C'}}^{x'_{C'}} 2z(x) dx' \quad (28)$$

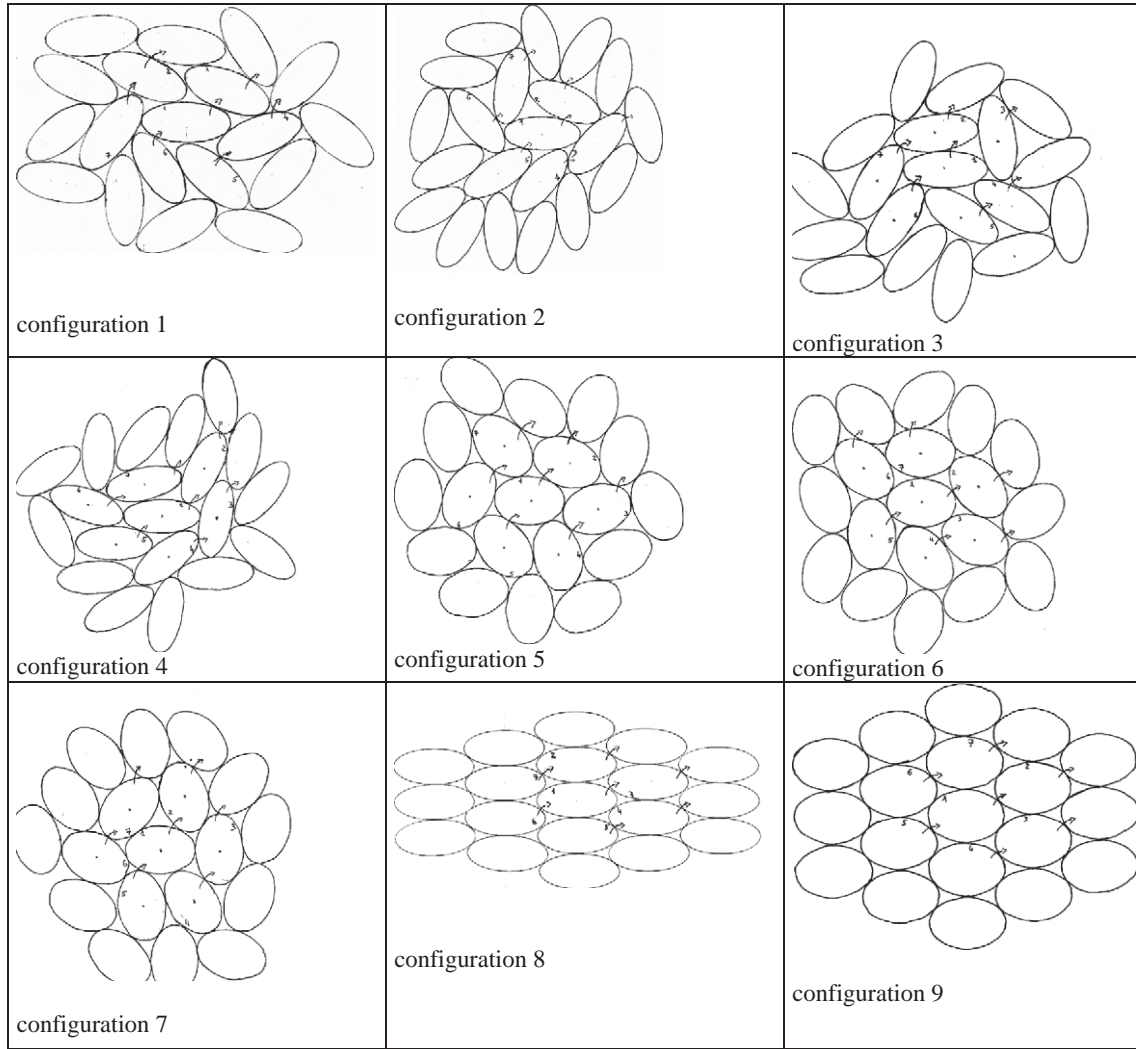


Fig. 10. Configurations of ellipsoids of revolution used to calculate x_u (top view).

and then

$$O'H = \sqrt{(x_H - x_O)^2 + (y_H - y_O)^2} \quad (34)$$

the solid angle is equal to:

$$\omega = \pi(1 - \cos\gamma) \quad (35)$$

This solid angle is half the classic one (cf. Eq. (1)) because in our case, we only consider the upper half space.

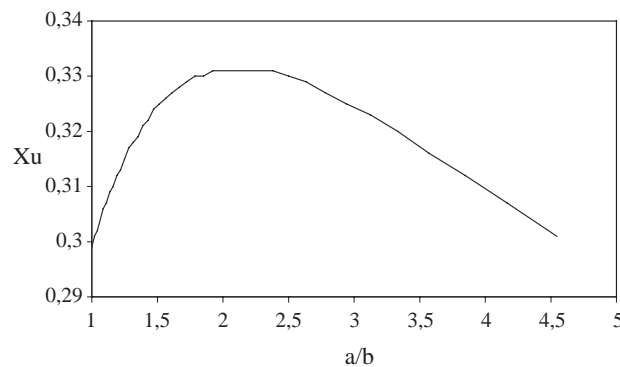


Fig. 11. x_u as a function of the major axis/minor axis (a/b) ratio of the ellipsoids.

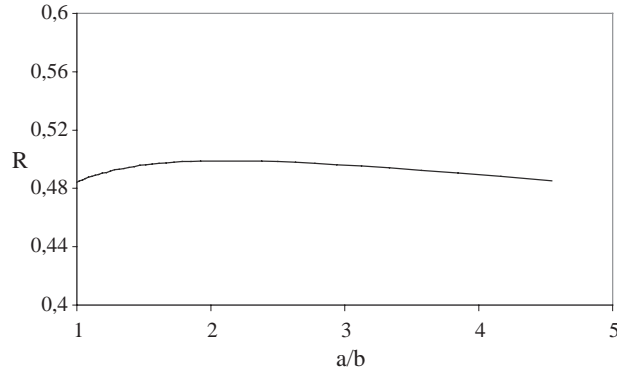


Fig. 12. Reflectance value as a function of the long axis/short axis (a/b) ratio of the ellipsoids.

4.3.2. Calculation of x_u

To calculate x_u , we have to sum the solid angles occupied by each neighbouring particle. But if we add without some care, some parts of the space will be accounted for two times, because, in some cases, a part of the surface we calculate for the ellipse i is masked by an ellipse j , situated between the ellipse i and the central ellipse (Fig. 9). In that case, it is necessary to make a correction.

In the case presented in Fig. 9, it is necessary to get the co-ordinates of B' . B' is the intersection between the straight line defined by the two tangential points to the ellipse i ($B_i C_i$) and one of the two tangent lines to the ellipse j , here the one passing through C_j .

To get the co-ordinates, it is necessary to get the co-ordinates of C_j in the system defined on the ellipse i . That can be done knowing the geometrical relations between the two ellipses (angle between the axis, co-ordinates of the centre in the new system). Then we can continue the calculation as before. Sometimes two corrections are necessary, the second is treated in the same way.

4.3.3. Calculation code

A numerical code has been written in order to calculate x_u for several different configurations taking into account all the geometrical considerations described before.

5. Results

Table 1 shows the variations of x_u for randomly drawn configurations, and for two different shapes of ellipsoid of revolution characterised by the rate a/b . Fig. 10 shows all the configurations used.

We note that, as we take more particles into account to calculate x_u , the variations between configurations become less important, particularly when the shape is spheroidal (a/b close to 1). So we can suppose that the variations encountered in large, semi-infinite samples are even weaker because far more particles are taken into account when a reflectance measurement is done. So that is why we choose the simplest configuration (configurations 8 and 9 in Fig. 10), to see the influence of particle shape on x_u .

This approximation is easily justified for ellipsoidal rather spherical particles, but it seems that x_u is minimised for rod shaped particles.

Fig. 11 shows the variations of x_u as a function of the a/b ratio in this simplest configuration. Fig. 12 shows the evolution of the reflectance value as a function of a/b , all the other parameters being kept constant.

We notice that the differences of x_u seem to be important in Fig. 11, but Fig. 12 shows that these differences have a weak influence on the calculated reflectance, which is our meaningful physical parameter. We therefore decided to keep the initial value of 0.299 (which we round up to 0.3) for x_u . Even so, we must not forget that this approximate value is probably too low for rod shaped particles.

6. Conclusion

This study shows that a coefficient, which can be quite different for individual particles, can be “globalised” if we take into account more particles, and then, we see that it becomes less varying. This globalisation is justified because in our problem, the property we want to be able to calculate (i.e. reflectance of the powder) is not the individual property of one particle but a collective property, which is the result of the assembly of all the particles. It is not important to know the colour of one isolated particle because there is experimental evidence that this colour is modified by neighbouring particles.

The conditions on the size and shape of the particles were an important limitation for the use of this model. This study allowed us to prove that the model can be used for industrial powders having a major axis/minor axis ratio as high as 4.5, which is the case of a large majority of industrial powder to continue to adapt the model for use with industrial powders. Results and applications are presented in [6].

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