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The effect of the particle shape and structure on the flowability of electrolytic copper powder. I. Modeling of a representative powder particle

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Abstract: One of the most important properties of copper powder is its flowability which depends on the shape and the structure of the powder particles. A procedure for the determination of a representative powder particle permitting the free flow of copper powder is proposed.

Keywords: copper powder flowability, representative particle of flowing powder, surface structure of particles of flowing powder.

INTRODUCTION

The flowability of a copper powder depends on the interparticle friction, which is dominated by the surface area and surface roughness of the particles. As the surface area and surface roughness increase, the amount of friction in the powder mass increases and the powder exhibits less efficient flow. The same appears with the shape of particle. The more irregular the particles shape is, the less efficient is the powder flow. Resistance to flow is the main feature of friction, which decreases as the particles approach a smooth spherical shape. The effect of particle size distribution on the powder flowability is also important. If the powder consists of monosized particles, which are more or less in point contact with one another, making the contact surface as low as possible, even dendritic deposits can flow. If the powder consists of different particles, the interstitial voids of the larger particles can be filled by the smaller ones, the contact surface area increases, and the flow of the powder is less efficient.[1] As a result of this, a non-sieved powder often does not flow, while the fractions of the same powder flow.[2,3] Hence, the best conditions for the free flow of the powder are fulfilled if the powder consists of monosized particles of spherical shape with a surface structure approaching the structure of a smooth metal surface. The aim of this work was to extend the recently developed concept of

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the representative powder particle on the flowability of a powder,[4] by finding the surface structure of a particle which permits the free flow of the powder.

DISCUSSION

In a previous work[5] it was assumed that free flow of a powder which consists of spherical particles can be expected only if the surface parts of the particles, corresponding to the metal segments, are larger than, assuming uniform distribution, or equal to the pores between them. Aside from this, free flow of a metal powder also depends on the surface structure of the particles. This can be confirmed by several examples.

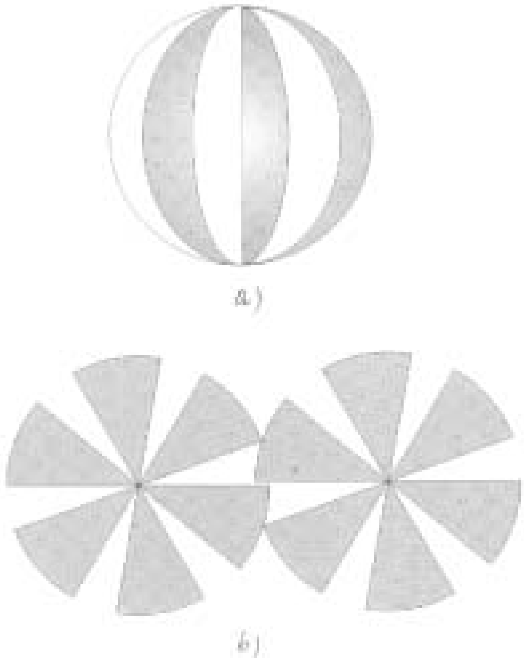


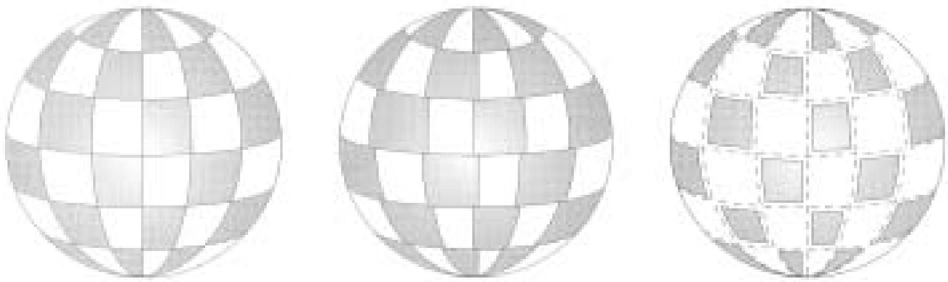
Fig. 1. The model of slices: a) overview; b) cross-section of the model of slices; position when jamming is possible.

A model which represents a sphere divided into slices of equal dimensions is presented in Fig. 1. Every second slice actually represents empty space, providing the volume of full slices is equal to the volume of empty ones. At the first sight, this model does not allow jamming during flowing, because of the equal size, the full segments will not be able to enter into the empty ones. However, if one pays attention to Fig. 1b, it can be noticed that the slices, owing to free corners, will be able to stick into vacations between them, even if the latter ones would be of much smaller size (in Fig. 1b the ratio of the volumes of the full to the empty segments is 2:1). This means that, apart from the segment size, their mutual arrangement also plays an important role in powder flowability.

Metal powders can also be presented by a model, which is reminiscent of the planet Earth. Namely, this model represents a sphere segmented by vertical and horizontal lines, in the same way the Earth is divided by meridians and parallels. It is considered that after every full segment follows an empty segment both in the horizontal and vertical direction. This means that every full segment on the surface of the sphere is surrounded on all sides by empty spaces (Fig. 2a). In order to achieve such a structure, it is necessary to have an even number of "meridians" (vertical lines) that gives an even number of slices (vertical segments) and an odd number of "parallels", that again gives an even number of layers (horizontal segments).

It can be assumed that the model is composed of slices of equal dimensions and layers providing for arcs of equal length (in the vertical direction) on the surface of the sphere. This means that all segments in one layer are of the same shape and size. Like in the previous case, the main characteristic of this model is that, regardless of the dimensions of the sphere and the number of its segments, the volume of the full segments will be always equal the volume of the empty ones. This implies that the volume of this model is exactly half of the volume of a full sphere of equal dimensions.

Another feature that this model provides is free flow. In order to have flowability, it is necessary for a powder to be composed of such particles in which the full segments will not be able to become stuck into the empty spaces of other particles. At first sight it seems that the model presented in Fig. 2a will not be appropriate for free flow. Namely, all the segments in a layer are of the same size, which gives the possibility of full segments entering into empty ones. Besides, in the other layers are segments of different dimensions (and shape in the last one), that additionally reinforces the expectation concerning jamming.



a) b) c)

Fig. 2. The Earth model: a) the surface of the full segments is equal to the "surface" of the vacancies; b) the surface of the full segments is larger than the "surface" of the vacancies; c) the surface of the full segments is smaller than the "surface" of the vacancies.

However, if the model presented in Fig. 2a is considered, it can be noticed that every full segment at all of its corners is connected with other ones, generating some kind of net on the surface of the sphere. Owing to this structure, where no segment is free and so not able to enter even much larger holes, it can be considered that jamming will be avoided and, consequently, the powder will be free flowing. The same conclusions can be made for the model where full segments are larger then the empty ones (Fig. 2b). It is obvious that jamming is impossible and that the powder will flow.

Now, the situation opposite to that in Fig. 2b can be assumed. A model is presented in Fig. 2c where the full segments are smaller then the empty ones. Here it is obvious that the full segments will be able to enter the vacancies between them. The reason for this is not just their smaller size, but also the fact that they are now not connected to other full segments, such as was the case with the model in Fig. 1.

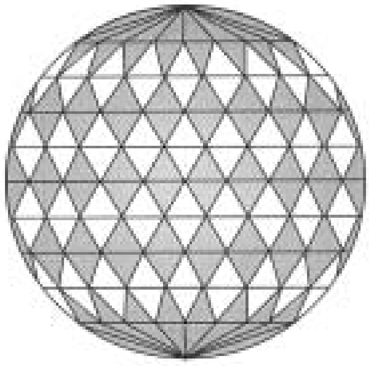


Fig. 3. The model of rhombuses.

The following model gives further evidence that the surface structure, alongside the segment and pore size, plays a crucial role for free flow. This model (Fig. 3) represents a sphere, where, in contrast to the previous model, all the segments are of the same shape, having triangles as the basic unit. Both semi-spheres are divided by parallel circles into an odd number of layers with equal arc lengths on the surface of the sphere. In order to achieve this, it is necessary for all parallel circles to have equal central angles, so each semi-sphere is divided itno $2n + 1$ ($n = 1, 2, ...$) identical angles. Then, every layer is segmented into an equal number of triangles and the space between them is considered as empty. Triangles in two neighbouring layers are connected mutually through their base, giving the shape of rhombus. Bearing in mind that the length of the triangle base depends on the size of the parallel circles, it can be noticed that, on moving towards the poles, the size of the rhombuses becomes smaller and smaller. It can be considered that these rhombuses (full segments) of different size represent, actually, different branches of the metal powder particle.

Like in the previous model, in this case it is also obvious that the surface structure is suitable for free flow. All segments at their corners are connected with others, so jamming will be impossible. However, from this model, it is not clear whether the volume of the full segments is equal to the volume of the vacancies, since full and empty triangles in the same layer are not of the same size. As a result of this, it is necessary to show that these two volumes can be considered as equal.

Now, spherical coordinates defined by the equations: $x = r \cos\varphi \sin\theta$; $y = r \sin\varphi \sin\theta$; $z = r \cos\theta$; are introduced.

The geometric meanings of r, φ and θ are shown in Fig. 4a. If the Jacobian of this mapping is denotet by $J(r, \varphi, \theta)$ then $|J| = r^2 \sin\theta$.

The volume of the first solid (the part composed of all full parts in layer 1) is equal to (see Fig. 4b):

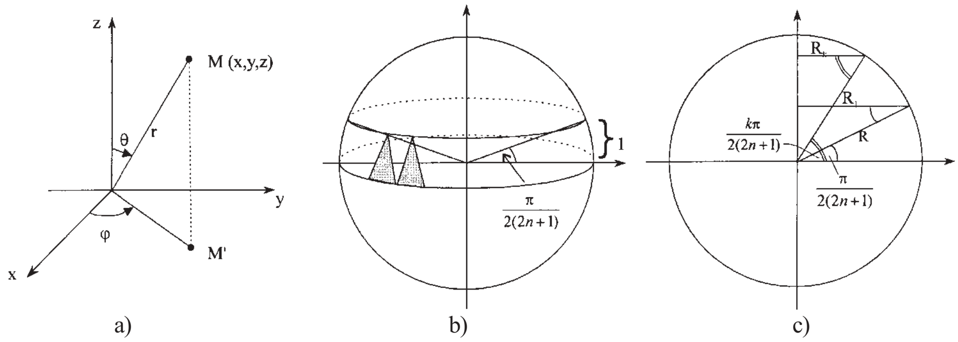


Fig. 4. Geometric interpretation: a) the spherical coordinates; b) geometric characteristics of the first layer; c) the ratio of R_k/R.

$$V_t = \int_0^{2\pi} d\varphi \int_{\frac{\pi}{2}-\frac{\pi}{2(n-1)}}^{\frac{\pi}{2}} \sin\theta d\theta \int_0^R r^2 dr = \frac{2\pi R^3}{3} \sin\frac{\pi}{2(n+1)} \tag{1}$$

Similarly, the volume of the k^{th} solid is equal to

$$V_k = \frac{4\pi R^3}{3} \cos\frac{(2k-1)\pi}{4(2n+1)} \sin\frac{\pi}{4(2n+1)} \tag{2}$$

Later, if the volumes corresponding to full and empty parts of the first solid are denoted by $V_1^{(F)}$ and $V_1^{(E)}$, respectively, then the ratio $V_1^{(F)} / V_1^{(E)}$ is equal to the ratio of the parameters of the boundary circles of that solid, *i.e.* of the ratio of their radii (Fig. 4c).

$$\frac{R}{R_1} = \frac{1}{\cos\frac{\pi}{2(2n+1)}}$$

Hence

$$\frac{V_1^{(F)}}{V_1^{(E)}} = \frac{1}{\cos\frac{\pi}{2(2n+1)}}$$

and since $V_1^{(F)} + V_1^{(E)} = V_1$, then by using (1) it can easily be shown that

$$V_1^{(E)} = \frac{2\pi R^3}{3} \tan\frac{\pi}{4(2n+1)} \cos\frac{\pi}{2(2n+1)} \tag{3}$$

and so

$$V_1^{(F)} = \frac{2\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \tag{4}$$

By using the same idea as in previous case, the following formulas for the k^{th} solid can be derived

$$V_k^{(*)} = \frac{2\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \cos \frac{k\pi}{2(2n+1)} \tag{5}$$

$$V_k^{(**)} = \frac{2n\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \cos \frac{(k-1)\pi}{2(2n+1)}, \qquad k = 1,2, ..., 2n+1 \tag{6}$$

where for odd k instead of * in Eq. (5), F is used and instead of ** in Eq. (6), E is used and the contrary for even k.

The sum of all the E-volumes for the semi-sphere is equal to

$$\sum V_k^{(E)} = \frac{4\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \left(\cos \frac{2\pi}{2(2n+1)} + \cos \frac{3\pi}{2(2n+1)} + \ldots + \cos \frac{2(n-1)\pi}{2(2n+1)} \right) =$$

$$= \frac{4\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \cdot \frac{\sin \frac{n\pi}{2n+1)}}{\sin \frac{\pi}{2(2n+1)}} \tag{7}$$

and for all the F-volumes

$$\sum V_k^{(F)} = \frac{4\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \left(\frac{1}{2} + \cos \frac{2\pi}{2(2n+1)} + \cos \frac{4\pi}{2(2n+1)} + \ldots + \cos \frac{2n\pi}{2(2n+1)} \right)$$

$$= \frac{4\pi R^3}{3} \tan \frac{\pi}{4(2n+1)} \cdot \frac{1}{\sin \frac{\pi}{2(2n+1)}} \tag{8}$$

It should be noted that in Eq. (7) and Eq. (8), the following results were used

$$\cos x + \cos 3x + ... + \cos (2n-1)x = \frac{\sin 2nx}{2 \sin x}$$

and

$$\frac{1}{2} + \cos 2x + \cos 4x + ... + \cos 2nx = \frac{\sin(2n+1)x}{2 \sin x} \qquad x \neq l\pi,\ l \text{ integer}$$

The ratio of those sums for a semi-sphere as in a sphere is equal to

$$\frac{\sum V_k^{(E)}}{\sum V_k^{(F)}} = \sin\frac{n\pi}{2n+1} = \sin\frac{\pi}{2n+\frac{1}{n}} \rightarrow \sin\frac{\pi}{2} = 1 \tag{9}$$

when $n \rightarrow \infty$, being lower for $0 < n < \infty$.

Bearing in mind that the volume of a segment actually depends on the product of its base area and its altitude (equal to R) and that all radii R in the sphere are of the same size, it can be concluded that the ratio of the surface area of these two kinds of segments is equal to the ratio of their volumes. This conclusion can be of great importance if it can be extended, even approximatelly, to non-spherical particles.

By decreasing the size (and simultaneously the volume, of the full segments, their corners will become free and they will be able to jam into empty spaces. This indicates that free flow will be possible in cases when the volume of the full segments is equal or larger then the volume of the pores, when the surface of the powder particles behaves litle those of smooth massive metal. The direct illustration of the above conclusions will be the aim of part II of this paper.

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ИЗВОД

УТИЦАЈ ОБЛИКА И СТРУКТУРЕ ЧЕСТИЦЕ НА ТЕЧЉИВОСТ ЕЛЕКТРОЛИТИЧКОГ БАКАРНОГ ПРАХА. I. МОДЕЛОВАЊЕ РЕПРЕЗЕНТАТИВНЕ ЧЕСТИЦЕ ПРАХА

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Једно од најважнијих својстава бакарног праха је његова течљивост, која зависи од облика и структуре честица праха. Предложен је поступак за одређивање репрезентативне честице праха која обезбеђује слободно течење бакарног праха.

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