The critical size of brittle solid materials and the measurement of their surface tension

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Abstract

The relationship of the energy required to break a particle or a particulate material vs. particle size has been studied by many researchers. On the one hand, mineral processing engineers, who are interested in the specific energy (in joules per cubic meter or joules per kilogram) required for grinding, almost agree that it is inversely proportional to the particle size, although they might disagree on the type of the relationship. On the other hand, building and structural engineers, who are mainly interested in the strength of materials (in newtons per square meter or pascals), they almost agree that at the size range of the elements used, their strength depends on the quality of the material rather than its size. The present article shows that both groups of engineers are right about the size range of the bodies used by each one. However, there is a critical size that determines the fracture behavior of a brittle material. The definition of the critical size can be used to understand the practical results obtained as well as to measure the surface tension of the tested materials.

Keywords: critical size; energy size relationship; grinding; specific energy.

1. Introduction

In laboratory tests for the strength of brittle materials, under uniaxial loading, the specimens have geometrical shapes (usually cubes or cylinders) with predetermined dimensions. The parameters measured are the force per unit area, also called stress, the absolute or relative deformation, and the maximum stress under which the specimen collapses. The unit of measurement is force per unit surface area, and in the MKS system, it is newtons per square meter (N/m^2 , also symbolized by Pa, or pascal). It is important to remind here that 1 Pa is equivalent to 1 J/m^3 , as $\text{Pa} = \text{N/m}^2 = \text{N} \cdot \text{m} / \text{m}^2 \cdot \text{m} = \text{J/m}^3$. The concept of specific energy, energy per unit volume, is more familiar in mineral comminution, crushing, and grinding, where the energy consumed to reduce the size of a material is more important than the stress applied to break a single particle.

The energy required to break a single particle has been studied by many researchers in the past, but we shall concentrate on two characteristic works by Kick [1] and Tavares

and King [2]. Kick [1] worked with small spheres of pig iron, and his conclusion is that the energy (E_k) required to break a particle is proportional to its volume (V)

$$E_k = e_k \cdot V \quad (1)$$

or to its mass (m), as $m = V \cdot \rho$, where ρ is the density of the material. The proportionality coefficient (e_k) is the volume-specific energy of the material that is found constant in the size range of spheres tested by Kick [1], i.e.,

$$e_k = \frac{E_k}{V} = \text{constant} \quad (2)$$

Tavares and King [2] have used a more sophisticated technique of dropping weight that has enabled them to test mineral particles smaller than 1 mm. Their findings are illustrated in Figure 1. Here one can see that, for relatively large particles, the volume-specific energy is constant, whereas for lower sizes, the specific energy increases inversely proportional to the size. In the log-log graph used, the inversely increasing part of the curve is a straight line with a more or less same slope regardless of the material used. The horizontal part of the curves corresponds to the specific energy of the tested materials and is in agreement with Kick [1]. It shows the internal cohesion energy that has to be overcome to break a particle and depends on its material.

In mineral processing where the measured parameter is the specific energy consumed to reduce the size of a material, it is always found to be inversely proportional to the particle size. The size range of particles involved in a grinding process is below that of the specimens used for measuring the strength of materials in uniaxial loading tests. There are many expressions that give the energy size relationship in grinding mills, but it has been understood that the energy consumed by comminution equipment is much greater than the actual energy consumed for breaking the particles of the material. Consequently, the proposed relationships give an overall energy consumption that involves mill performance as well.

The theory that seems to have a theoretical value is that of Rittinger [3] and relates the pure energy (E_r) consumed with the new surface (ΔS) produced when coarse particles break to produce smaller ones. The relationship can be written in the following form

$$E_r = \gamma \cdot \Delta S. \quad (3)$$

The coefficient γ is the surface tension of the material defined as the energy required for producing a unit of new surface area. The specific energy according to Rittinger [3] is

Figure 1 Specific energy-size relationship by Tavares and King [2].

given by Eq. (4) and depends on the ratio of surface area to the volume that is geometrically size dependent, i.e.,

$$e_R = \frac{E_R}{V} = \gamma \cdot \frac{\Delta S}{V} \quad (4)$$

The present article proposes a model to explain the energy size relationship experienced during fracture tests as well as a simple method of measuring the surface tension of brittle materials.

2. Synthesis of the model

From geometry, one has that the volume of a sphere of diameter (d) is

$$V = \frac{\pi}{6} d^3 \quad (5)$$

According to Kick [1], the energy of a dropping weight just enough to break the sphere is given by Eq. (6), calculated by substituting Eq. (5) into Eq. (1), and statistically, it produces two hemispheres.

$$E_K = e_K \cdot \frac{\pi}{6} d^3. \quad (6)$$

Solving for the specific energy as defined by Kick one has that it is constant independent of the size, i.e.,

$$e_K = \frac{6 \cdot E_K}{\pi \cdot d^3} = \text{constant}. \quad (7)$$

The value of e_K is easily determined experimentally for relatively large particles because one can always calculate the minimum energy of the dropping weight as well as the mass, volume, and diameter of the particle.

After fracture, the total surface area of the two hemispheres is increased compared with the initial sphere, and the increment (ΔS) is equal to the two new surfaces formed. From geometry, one has

$$\Delta S = \frac{\pi}{2} d^2. \quad (8)$$

Substituting Eqs. (8) and (5) into Eq. (4), one can calculate, according to Rittinger [3], the minimum specific energy required to produce the two new surfaces, i.e.,

$$e_R = \frac{3 \cdot \gamma}{d}. \quad (9)$$

The difficulty in calculating the specific energy from Eq. (9) is that the value of the surface tension of solids is not easily determined. As will be discussed later, the methods proposed for the measurement of surface tension, such as the Brazilian test described by Kaklis [4], Colback [5], and Malan and Napier [6], ignore the critical size. On the other hand, the method described by Stamboliadis [7] requires tensile strength measurements that demand fibers of the material with varying fine diameters that can be practically obtained for glass but not for minerals.

Theoretically, for relatively large spherical particles, the specific energy required to overcome the cohesive strength of a material is usually greater than the energy required to produce the two new surfaces. Consequently, at large sizes, the controlling factor is the cohesion strength of the material that does not depend on the size. One exception could be materials with crystal deformations at the size range of the particles such as minerals with cleavage. However, according to Rittinger [3], because the specific energy to produce the new surfaces increases as the particle size is reduced, there must be a critical size (d_c) at which both specific energies defined by Eqs. (7) and (9) are equal. At particle sizes lower than the critical one, the controlling factor is the surface area formation that increases inversely proportional to the particle size. This critical particle size is found by equating (7) with (9) and solving for d

$$d_c = \frac{3 \cdot \gamma}{e_K}. \quad (10)$$

If d_c could be determined, the surface tension (γ) could also be calculated as well, thus solving Eq. (10) as

$$\gamma = \frac{e_K \cdot d_c}{3}. \quad (11)$$

In Eq. (11), the surface tension (γ) is calculated from the two well-defined parameters: e_K and d_c .

2.1. A theoretical example

Assume spheres of a material with specific energy $e_K = 600,000 \text{ J/m}^3$ (Pa) and $\gamma = 300 \text{ J/m}^2$ (N/m). Using Eq. (10), the calculated critical size is $d_c = 3 \times 300 / 600,000 = 1.5 \times 10^{-3} \text{ m}$

(1.5 mm). According to the above model, one can present the specific energy-size relationship in a diagram preferably in log-log scale. Taking logarithms in Eq. (2) and substituting the value of (e_K) , one obtains $\log(e_K)=\log(600,000)$ presented by line “Kick” in Figure 2. On the other hand, taking logarithms in Eq. (9) and substituting the value of γ , one obtains $\log(e_R)=\log(3 \times 300)-\log(d)$ presented by line “Rittinger” in the same figure.

The two lines cross each other at the critical size (d_c). Above the critical size, e_K is greater than e_R and is constant, whereas below critical size, e_R is greater than e_K , and it increases continuously with decreasing size. The specific energy required to break a particle of any size is the sum of the two obstacles that have to be overcome and is also presented in Figure 2 by the thick line labeled total. At coarse sizes, one has to provide the energy required to overcome the cohesion forces of the material, and at fine sizes, the energy provided must be enough to create the new surfaces produced by fracture. At intermediate sizes, where both energies are of the same magnitude, the total line is curved. This is the type of curves obtained in practice by Tavares and King [2] (presented in Figure 1).

2.2. Actual measurement of surface tension

Once the model has been presented, one can use it for the actual calculation of surface tension from real experiments. The data used are that presented so far in Figure 1, namely, the data for marble and taconite. Although the exact arithmetic data are not available, one can have a close estimate from Figure 1. For marble, the estimate is $e_K=0.12 \text{ J/cm}^3$ ($120,000 \text{ J/m}^3$), the “Kick” and “Rittinger” lines cross at the critical size $d_c=1.2 \text{ mm}$ (0.0012 m). Using Eq. (11), one has $\gamma_{\text{marble}}=48 \text{ J/m}^2$. Following the same procedure for taconite, one has $e_K=600,000 \text{ J/m}^3$, $d_c=1.4 \text{ mm}$ (0.0014 m), and the surface tension is calculated at $\gamma_{\text{taconite}}=280 \text{ J/m}^2$.

3. Discussion

The type of curves obtained when measuring the volume-specific energy of brittle materials as a function of particle size (presented in Figure 1) has already been explained

Figure 2 Theoretical specific energy-size relationship.

theoretically using the concepts of the cohesion energy and surface tension of materials [7]. In that case, the problem is approached from the point of view of the deformation taking place when a specimen is under load. According to this approach, the energy provided is partly stored in the specimen as internal energy and partly consumed to increase the surface area of the specimen as a result of its deformation. The particle breaks when the energy stored as internal exceeds the cohesion energy per unit volume of the material. The total energy consumed is the sum of the internal one and the one used for surface deformation up to the point of rupture. For large particles, the surface deformation energy is small compared with the cohesion energy, and the strength of the material is independent of size. At lower particle sizes, the surface deformation energy is greater than the cohesion energy and determines the apparent strength of materials. The already mentioned theory by Stamboliadis [7] uses Young's modulus to explain the phenomenon and does not immediately determine the critical size. In addition, it cannot be used for the measurement of the surface tension of materials under load but only in tensile strength experiments. The process of measuring the surface tension under tensile stress requires fine fibers of the material that are practically impossible to prepare for most types of materials such as minerals, although they can be prepared for materials such as glass.

The present model defines the critical size of a material, and this definition enables one to use common energy-size fracture techniques to measure the surface tension of solids. Let us consider other techniques used for the surface tension of solids such as the case of Brazilian test, already mentioned above. The experimental configuration is presented in Figure 3A, and the specimen is a disk of diameter (d) and thickness (t).

The volume (V) of the disk is $V=\frac{\pi \cdot d^2 \cdot t}{4}$. The energy (E_B)

measured to break the specimen into halves is equated with the energy (E_S) required to create the area of the two new surfaces, i.e.,

$$E_B=E_S \quad (12)$$

By definition, $E_S=\gamma \cdot \Delta S$, where $\Delta S=2 \cdot d \cdot t$, and consequently,

$$E_B=2 \cdot \gamma \cdot d \cdot t \quad (13)$$

Dividing by the volume, one obtains the specific energy (e_B) that, according to Kick [1], is constant

$$e_B=\frac{8 \cdot \gamma}{\pi \cdot d} \quad (14)$$

Figure 3 Brazilian test configuration.

Solving for γ , one obtains

$$\gamma = \frac{\pi \cdot e_B}{8} \cdot d \quad \text{or} \quad \gamma = k \cdot e_B \cdot d \quad (15)$$

where k is the shape parameter. As e_B is constant, the result obtained as the value of γ is not constant and depends on the size of the specimen. Of course, this is not correct because the surface tension is a property of the material independent of the size, unless there are defects of the material in the size of the specimen tested. The source of mistake is Eq. (12), which, as explained in the text, is only true for the critical size (d_c). Because the size of specimens used for the Brazilian test is of the order of centimeters, whereas the critical size is of the order of millimeters, it is expected that under these conditions $E_B > E_S$, and consequently, the values of γ calculated are greater than the real ones. It is the author's opinion that the indiscriminant use of the size (d) of the specimen in calculating the surface tension (γ) is a conceptual mistake.

One could argue that the actual volume of the specimen affected by the stress is limited to a width (w) [4] (Figure 3B). In such a case, the volume is $V = d \cdot w \cdot t$, and Eq. (15) becomes

$$\gamma = \frac{e_B}{2} \cdot d \quad \text{or} \quad \gamma = k' \cdot e_B \cdot w \quad (16)$$

Here again, the value of γ depends on a size factor (w), which, by proportionality, is also a function of d . It is supported here that the size factors in Eq. (15) or Eq. (16) must be replaced by the critical size (d_c), which cannot be determined by the Brazilian test alone.

In mineral processing, the models or functions used to express the energy-size relationship are treated as if they were contradictory, especially the one attributed to Kick [1] compared with those proposed by Rittinger [3] or other researchers. There are several references to this contradiction by Stamboliadis [8, 9] that clarify the concept and show that the principles of Kick [1] and Rittinger [3] are not really contradictory. The present work is a further step to emphasize these principles and help to understand the conditions for their application.

In grinding, the formula most widely used to express the energy size relationship is Eq. (17), where d_1 is the initial size, d_2 is the final size of the material, and k is a constant, i.e.,

$$e = k \cdot \left(\frac{1}{d_2^n} - \frac{1}{d_1^n} \right). \quad (17)$$

According to Rittinger [3], the exponent $n=1$ because the energy is proportional to the new surface produced during

grinding and it also corresponds to the slope of the Rittinger line in Figure 2. According to Bond [10], $n=0.5$, whereas Charles [11] and Stamboliadis [12, 13] suggest that it can vary. Recently, Stamboliadis et al. [14] have proposed a method to calculate exponent n , using the same technique that Bond used to determine the work index of the materials. Hukki [15] proposed a Solomonic explanation to the problem. Based on the curved part of the total line in Figure 2, it is now clear that all these theories are only partially correct. It is also noteworthy that for almost all the minerals tested by Tavares and King [2] (presented in Figure 1), the slope of the curves at fine particle sizes is more or less constant equal to -1, which corresponds to Rittinger's theory.

Finally, the present work raises a question regarding the properties of a material that affects its fracture behavior. It is mentioned above that the behavior of brittle solids has been studied from the point of view of the deformation experienced during loading, and it was necessary to use three different properties, the Young's modulus (Y), surface tension (γ), and internal cohesion (e_K), to explain the behavior of the material. It was also noticed that what is now defined as the critical size was affected by changing either the Young's modulus (Y) or the surface tension (γ). The question or the challenge is whether there is a relationship among the three properties (Y , γ , and e_K) of the material or can they vary independently?

References

- [1] Kick F. *Das Gesetz der proportionalen Widerstande und seine Anwendung*. Arthur Felix: Leipzig, 1885.
- [2] Tavares LM, King RP. *Int. J. Miner. Process.* 1998, 54, 1–28.
- [3] Rittinger PR. *Lehrbuch der Aufbereitungskunde*. Ernst & Kern: Berlin, 1867.
- [4] Kaklis CN. *Characterization of Rock Anisotropy and Tensile Strength Using the Brazilian Test*, PhD thesis. Technical University of Crete: Chania, Greece, 2003, pp. 122–125.
- [5] Colback PSB. *Proc. First Congress International Society of Rock Mechanics*, 1967, pp. 385–391.
- [6] Malan DF, Napier JAL. *Int. J. Rock Mech. Min. Sci. Geomech. Abstr.*, 1994, 31, 581–596.
- [7] Stamboliadis ETh. *J. Mech. Behav. Mater.* 2005, 16, 363–377.
- [8] Stamboliadis ETh. *Can. Metall. Q.*, 2004, 43, 249–258.
- [9] Stamboliadis ETh. *Miner. Eng.* 2007, 20, 140–145.
- [10] Bond FC. *Trans. AIME Min. Eng.* 1952, 193, 484–494.
- [11] Charles RJ. *Trans. AIME Min. Eng.* 1957, 208, 80–88.
- [12] Stamboliadis ETh. *Miner. Eng.* 2002, 15, 707–713.
- [13] Stamboliadis ETh. *EJMP&EP*. 2003, 3, 160–166.
- [14] Stamboliadis E, Emmanouilidis S, Petrakis E. *Geomaterials*. 2011, 1, 28–32.
- [15] Hukki RT. *Trans. AIME*. 1961, 220, 403–408.