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# Fracture properties of spray-dried powder compacts: Effect of granule size

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**Abstract**

Inappropriate mechanical properties of spray-dried powder compacts lead to significant green product losses, entailing considerable costs in ceramic tile manufacture as well as serious environmental problems. In addition, green strength can be indicative of how well a ceramic processing system is working.

In this study, granules were prepared by spray drying a red clay slurry used in floor tile manufacture. The resulting granules were characterised and their porosity, morphology, and mechanical behaviour were determined.

The study analyses the fracture properties of green ceramic materials using Linear Elastic Fracture Mechanics (LEFM), which has been widely used for fired materials, but whose application to green compacts has drawn much less attention. Two types of tests for determining fracture parameters (fracture toughness, fracture energy, and crack size) in green materials are also critically examined. Finally, the fracture parameters have been correlated to the microstructural characteristics of the compacts, in particular to granule size and the topography of the fracture.


*Keywords:* Pressing; Green strength; Clays

## 1. Introduction

Green mechanical strength is a key property in green pressed bodies, since it is essential these compacts should be able to withstand, without damage, the thermal and mechanical stresses they undergo during the pre-firing stages. In addition to green handling considerations, green strength can be indicative of how well a ceramic processing system is working [1–3]. Defects originating in the earliest processing stages have been shown to persist into the final product [4,5]. Current product losses from cracking and breakage of green tiles are estimated at 3%, while fired product losses from cracks in green tiles are estimated at about 2%. This means that in Europe alone, every year, the losses associated with tile cracks and breakage amount to about 200 million euros, in addition to some 625 000 tons of solid wastes that mainly go to landfills. Powder compact strength is also of interest from a scientific viewpoint, since it provides information on the packing structure and particle interaction forces. The strength of green bodies has thus been the subject of numerous studies. In order appropriately to explain a material's mechanical behaviour during fracture and to attempt to relate a material's mechanical strength to its microstructural characteristics, or to try to explain the role of ceramic processing in mechanical strength, it has often been necessary to use fracture theories such as Linear Elastic Fracture Mechanics (LEFM). Indeed once tensile strength, $\sigma_f$, and fracture toughness, $K_{IC}$, have been determined, critical crack size, $a$, can be calculated using LEFM. This approach is very interesting when the critical crack size can be related to processing conditions or microstructural characteristics. Few attempts have been made, however, to apply LEFM to failure in green bodies. For porous particulate solids with low volume fractions of polymeric binders, Adams et al. [6–8], and later Ennis et al. [9], obtained fracture toughness, $K_{IC}^{A}$, from the fracture stress of straight-notched bars, $\sigma$, by the three-point bend test (single-edge notched beam) SENB, using the following equation:

$$K_{IC}^{A} = Y\sigma(\bar{a})^{1/2} \quad (1)$$

where $\bar{a}$ is effective crack length and $Y$ is a function of $\bar{a}$, the geometry and loading of the body [10]. The value of $\bar{a}$ is

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given by the sum of notch depth $a_0$, and a quantity $a'$, obtained together with $K_{IC}^{A}$ by fitting the pairs of values ($\sigma$, $a_0$) to the equation:

$$K_{IC}^{A} = Y\sigma(a_0 + a')^{1/2} \quad (2)$$

For these researchers critical crack size, $a'$, does not represent a natural flaw size but a process zone ahead of the crack tip where microcracking occurs, since critical crack size, which is several times larger than particle size, is not sensitive to binder composition, porosity, or particle size.

Bortzmeyer et al. [11] determined the fracture toughness of a green zirconia body, also using three-point bend bars with a straight notch (SENB). To calculate fracture toughness, $K_{IC}^{B}$, they applied Eq. (2), assuming that notch depth, $a_0$, practically coincided with effective crack length, $\bar{a}$. This is an acceptable assumption when the critical crack size, $a'$, is much smaller than $a_0$.

Bortzmeyer calculated critical crack size, according to the most common procedure, from bending strength, $\sigma_f$, and the value of $K_{IC}^{B}$, from the following equation:

$$a = \left(\frac{K_{IC}}{Y\sigma_f}\right)^2 \quad (3)$$

The critical crack size, $a$ (about 200 μm) calculated in this fashion was much larger than the maximum pore size (about 0.2 μm), determined by mercury porosimetry, and depended on forming conditions. Thus, critical crack size, $a$, decreased as pressed compact density increased.

Lan et al. [12] determined the fracture toughness of spray-dried alumina powder compacts from the fracture work per unit cross-sectional area, $G_C$, and the elastic modulus, $E$, using the equation derived by Irwin:

$$K_{IC}^{I} = (EG_C)^{1/2} \quad (4)$$

$G_C$ was obtained by integrating the load–displacement curve of the unnotched bars tested at a very low cross-head displacement rate (≈20 μm min$^{-1}$), because the fracture behaviour of the green bars at this rate was non-catastrophic. Critical crack size, $a$, obtained from Eq. (3), was found to be independent of compaction pressure, compact relative density, and granule hardness. In addition, the critical crack size, $a$ (about 300 μm) was five or six times the granule size.

More recently, Zhang and Green [13] and Uppalapati and Green [14] determined the fracture toughness of spray-dried powder compacts by notched diametral compression tests. Surprisingly, the calculated critical crack sizes, $a$, obtained from Eq. (3), increased with increased compaction stress. Just as the other researchers, they found that the magnitudes of the calculated critical crack sizes were relatively large (about 1 mm), several times the granule size (about 200 μm). It is hypothesised that subcritical crack growth can occur prior to failure at sites where the voids and defects in the compact coalesce, resulting in the large critical crack size. However, the effect of pressing pressure on critical crack size is inexplicable.

The chevron-notched three-point bend test (CNB) is often used to measure the fracture toughness of brittle materials such as ceramics, since it is a relatively simple experimental procedure while, in brittle materials, use of the chevron-notched specimen alleviates the problem of introducing pre-cracks in the sample. In addition, it is possible to calculate fracture toughness without measuring crack length or the load point displacement, using only the maximum load value recorded during the test [15]. In addition, once the continuous load–deflection curves have been obtained, the fracture work can be calculated, since crack extension is always controlled. Chevron-notch geometry presents an increasingly larger crack front to the advancing crack, thus forcing the crack to extend in a stable manner over the complete area of the chevron notch. This requirement cannot be readily achieved by standard three-point beam bending tests (notched or unnotched). Furthermore, with the CNB procedure, fracture toughness is practically independent of notch width [16,17], in contrast to what occurs with the SENB test [11,18].

Despite the advantages of the CNB test, and though it has been considered appropriate for the determination of toughness in highly porous materials, it even being recommended for green bodies [19,20], the test has never been used to characterise spray-dried powder compacts. Similarly, a literature search found no study on the effect of granule size on the fracture properties of green powder compacts, even though this characteristic has a notable effect on green [21–23] and fired [24] mechanical properties. Moreover, an examination of the effect of granule size on green characteristics and properties becomes of even greater interest if it is taken into account that almost all studies are conducted with granules obtained in pilot spray-driers, the granules thus being smaller than those produced on an industrial scale. This study has sought, first, to apply LEFM to the failure of spray-dried pressed compacts, and to determine the effect of granule size on body fracture properties and the relation between these properties and the microstructural characteristics of the pieces. Secondly, a comparative study has been conducted of the three-point bend test with straight-notched bars (SENB), the most widely used procedure, and the chevron-notched three-point bend test (CNB), with a view to using the latter in subsequent studies.

## 2. Experimental

### *2.1. Materials and slurry preparation*

Slurries of stoneware floor tile compositions were obtained [21]. They were prepared in industrial discontinuous ball mills (SACMI MTD 380) by mixing red clays, wastewater, and deflocculant (tripolyphosphate–metasilicate 3:1 in weight). The water used was very hard; its specific electrical conductivity was 1980 μS/cm. In order to select the optimum solids and deflocculant content, flow curves were determined using a Bolin CS-50 rheometer. The flow curves of slurries with the optimum deflocculant content (0.75 wt%) and different volume fraction ($\varphi$) are shown as examples in Fig. 1.

In order to attempt to relate resulting granule characteristics, such as morphology and porosity, to the rheological behaviour of the slurries [25], the flow curves were fitted to the Casson equation