

# COMPACTION BEHAVIOR STUDY OF POWDER COMPOSED BY NANOPARTICLE AGGLOMERATES AND AGGREGATES

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**Abstract:** Ceramic powder compaction by pressing requires weak-agglomerated powders to obtain high dense compacts and consequently dense sintered ceramic body. In this work powders with different agglomerated and/or aggregated size distributions was compacted by uniaxial and isostatic pressing. The powder and compacts were characterized by mercury and nitrogen porosimetry, BET and SEM. Analysis of X-rays diffraction peaks was used to determine the particle size (diameter). The Scherrer method was used to obtain the estimated average nanoparticle size. The results showed that all aggregated and weak agglomerated powders were composed by nanoparticles. This study indicated that the nanoparticle aggregates and weak agglomerates were destroyed during the pressing stage conducting to a increase of the packing degree of the powder particles.

## Introduction

Over the past several years a number of techniques have been developed for the production of ceramic nanoparticles. In a majority of these techniques the focus has been to reduce the primary particle size to below 30 nm. However, the problems related to agglomeration and aggregation have not been adequately addressed<sup>[1-3]</sup>.

The agglomeration refers to adhesion of particles to each other due to the van der Waals forces of attraction, which is significantly larger in nanoparticles. Nevertheless, aggregation refers to particles being necked together. As a result, it is difficult to break down an aggregated of nanoparticles into the individual primary particles. In general it was found that ceramic nanoparticles are difficult to compact at room temperature. However, several researches defeats the purpose that in a large number of applications it is important not only to have a small primary particle size (< 30nm), but also the secondary particle size (aggregates < 100nm)<sup>[1-5]</sup>.

Chemical methods are frequently used to obtain nanoparticulated and homogeneous powders. Ceramic nanoparticles prepared by chemical methods have advantages over physical methods in some respects as low cost, higher productivity level and ease of maintenance. One of the most critical steps in these methods is the extraction of the liquid phase (generally water) from the solid precipitated, or drying of the precipitate<sup>[3-6]</sup>. The way adopted to dry the

precipitates influences on the amount of agglomerates and/or aggregates in the powder<sup>[7]</sup>. In this work, compaction of several powders mixtures dried by different methods and consequently with different amount of aggregates and/or agglomerates was studied.

## Experimental Procedure

Different agglomerated powders were obtained from zirconium and cerium hydroxides coprecipitated and: i) conventional dried at 110°C (ZCH-S), ii) freeze dried (ZCH-L), iii) dried by extraction of water with ethyl alcohol (ZCAE) and ii) by extraction of water with isopropyl alcohol (ZCAP) [1]. It was used a commercial powder (ZS2 from MEL - Magnesium Elektron Ltd), and named ZCO, to compare the powder compaction characteristics. After the dry step the powders were calcinated at 400°C in order to obtain the oxide mixtures. These powder was characterized by Hg and N<sub>2</sub> porosimetry and SEM. Analysis of X-rays diffraction peaks was used to determine the particle size (diameter). The Scherrer method was used on the breadth of individual peaks to obtain the estimated average particle size. Green bodies were uniaxial (at 40 MPa) and isostatic (at 300 MPa) pressed. In order to investigate the compaction behavior of the different powders were used Hg and N<sub>2</sub> porosimetry.

## Results and Discussion

The selection of the powder mixtures of zirconia-ceria to this study was based on the disposability of several powders with different morphological characteristics. Besides, in a further work, we intend to investigate the effect of the aggregation and agglomeration states of these powders on the sintering mechanisms.

The powder aggregation and/or agglomeration states can be observed in the Figs.1 to 3. The powder characteristics were affected by the way of removing the water during the drying step. The mixture of commercial powders showed agglomerates and aggregates. However the powders obtained from coprecipitated hydroxides showed a major amount of agglomerates, except for this one dried in the conventional way (ZCH-S). X-rays peaks measurements gave the average particle size lower than 50 nm.

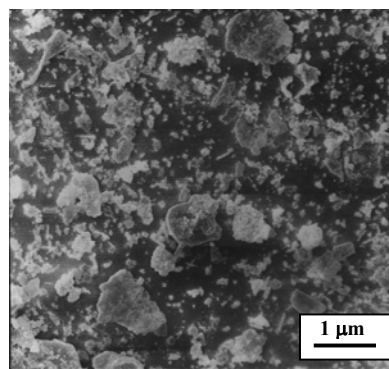


Figure 1 – SEM image of powder mixture of commercial oxides (ZCO).

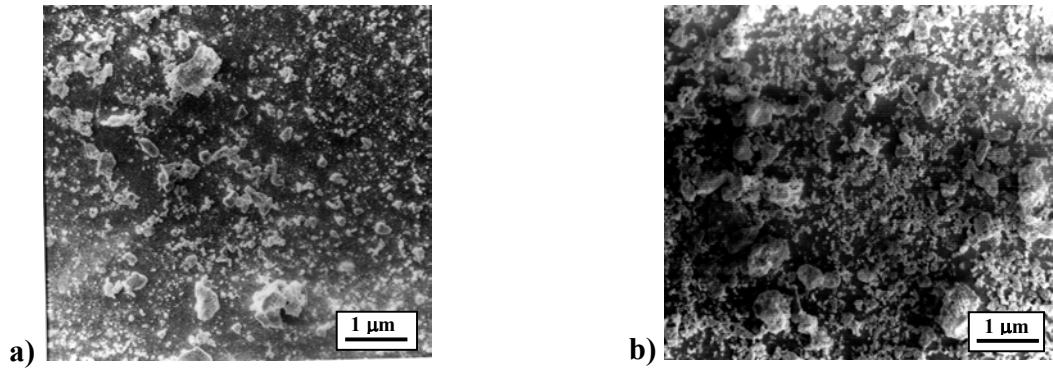


Figure 2 – SEM images of calcinated powder mixtures obtained from coprecipitated hydroxides and previously dried by: a) conventional method ZCH-S and b) freeze drying (ZCH-L).

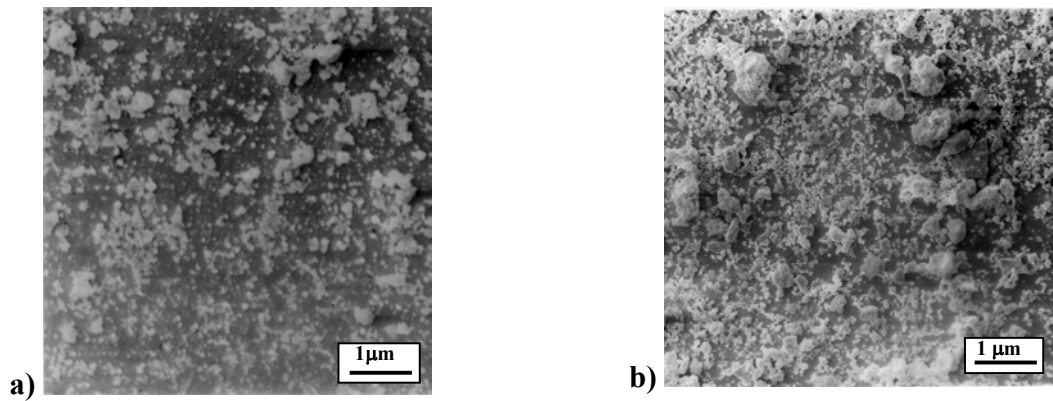


Figure 3 – SEM images of calcinated powder mixtures obtained from coprecipitated hydroxides and previously submitted to the water extraction by: a) ethyl alcohol (ZCAE) and b) isopropyl alcohol (ZCAP).

The curves of particle size distribution show the aggregates and/or agglomerates (Figs 4 and 5). However the agglomerates were destroyed during the sample preparation. This behavior

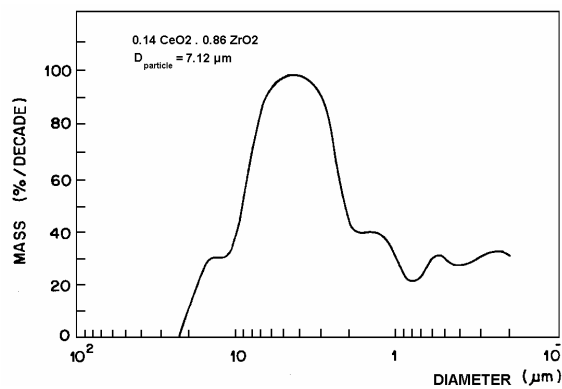


Figure 4 – Curve of particle size distribution for ZCO powder.

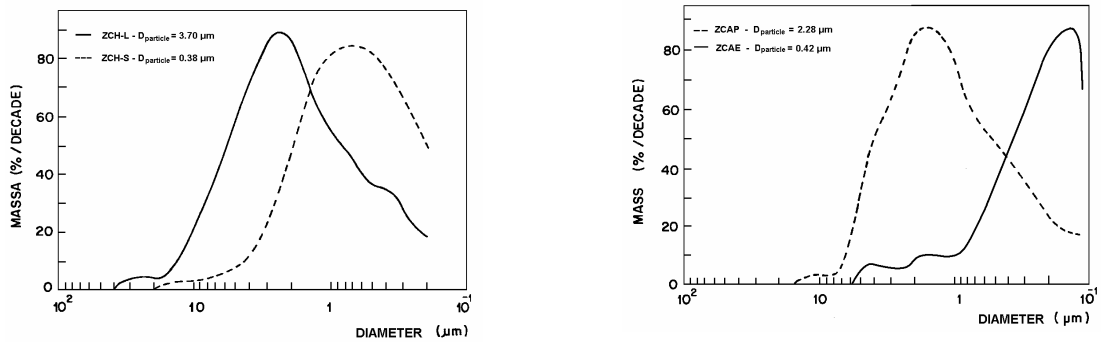


Figure 5 - Curves of particle size distribution for the calcinated powder mixtures obtained from coprecipitated hydroxides and previously dried by: a) conventional method (ZCH-S) and freeze drying (ZCH-L), and b) ethyl alcohol (ZCAE) and isopropyl alcohol (ZCAP).

was due to their weak particle-particle bonding. It is observed differences between these curves and the respective SEM images, due to the difficulty to prepare representative powder samples for microscopy observations. Therefore the informations from the curves of particle size distribution are insufficient to characterize the agglomeration state of the powder.

Pore size distributions for these powders showed a relative high pore volume for powders obtained from hydroxide precipitations (Fig. 6). During their densification these powders showed

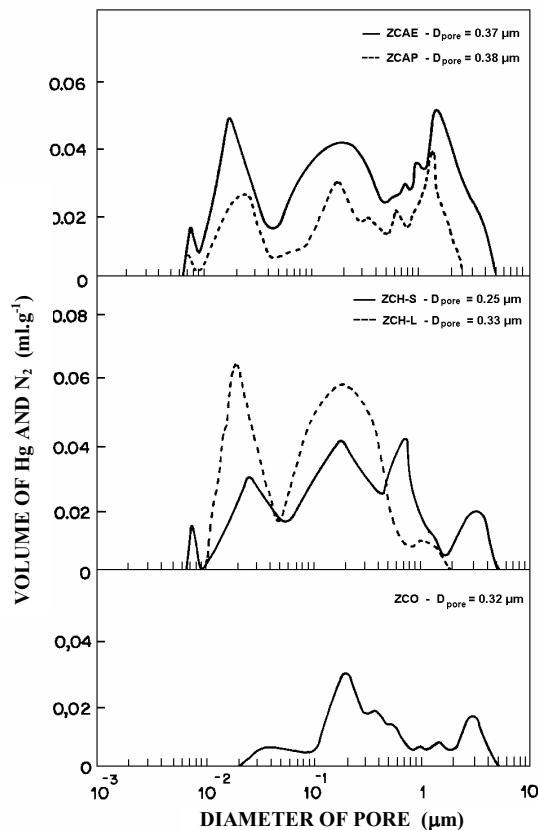


Figure 6 - Pore size distribution for the calcinated powder mixtures investigated in this work.

a high reduction on their pore volume. Powders obtained from hydroxide precipitations presented a decrease on volume proportional to the amount of agglomerates content. It is observed that powder that had aggregated nanoparticles showed pores larger than 100 nm in spherical

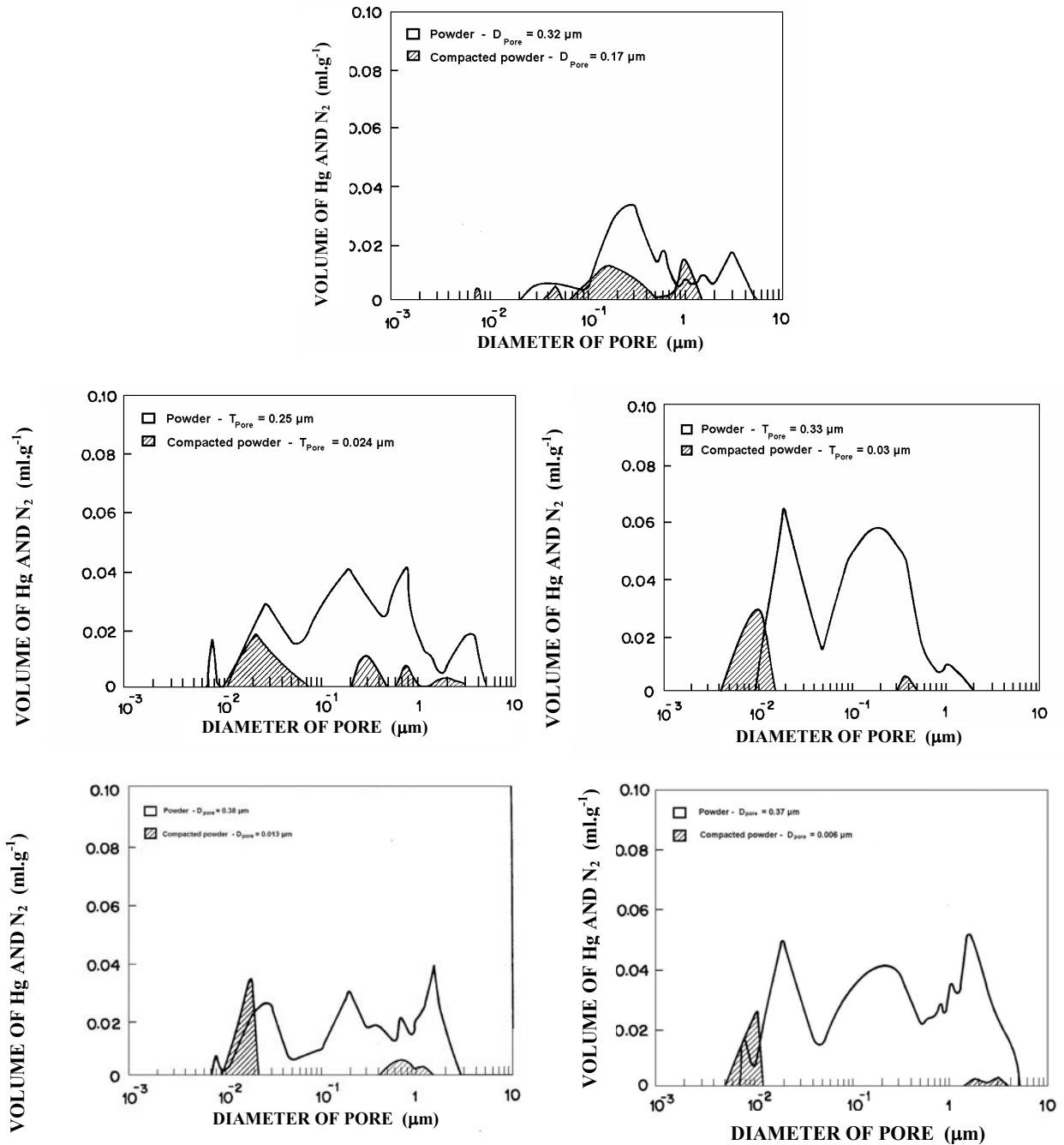


Figure 5 - Curves of particle size distribution for calcinated powder mixtures obtained from coprecipitated hydroxides and previously dried by different techniques.

equivalent diameter. Calcinated powder mixtures obtained from coprecipitated hydroxides and previously dried by freeze drying (ZCH-L) and water substitution using isopropyl alcohol (ZCAP) presented the best compaction degree. In these cases the majority of pores were found between 3 to 11 nm. However, in both samples there were small volumes of pore sizes between 0.2 to 4  $\mu\text{m}$  which related to packing flaws.

## **Conclusion**

The results showed that the agglomerates and aggregates in the powders studied were composed by nanoparticles (average particle size  $< 50\text{nm}$ ). Nanoparticle agglomerates were destroyed during the pressing stage conducting to an increase of the packing degree of the powder particles. This behavior was reported by other authors that used others characterization techniques of the green compacts. This study indicated that the combination of the porosimetry techniques and SEM supplies important information about the nanoparticle aggregation/agglomeration state and the capacity of accommodation of these particles during the compaction step (powder compaction ability).

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