

Study on Ceramic Membrane Support Sintering Oriented Preparation of Alumina Powder

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Abstract: Alumina powder is one of the most important raw materials of ceramic membrane support. The study is mainly about optimizing wet ball grinding conditions using alumina powder with size of 500 meshes as raw materials according to the needs of powder preparation, comparing grinding results on the different conditions including grinding time and the ball-to-powders ratio. Experimental results show that the best condition is that the ball-to-powders ratio of (9~11):1 and the best grinding time of 9~10 h. The subsequent experiments indicated that the powder milled on the best wet ball grinding conditions can be sintered directly into ceramic membrane support having high temperature resistance and high mechanical strength.

Keywords: Ceramic membrane support sintering, grinding time, preparation of alumina powder, the ball-to-powders ratio, wet ball grinding

INTRODUCTION

Membrane separation technology and its application developed rapidly in recent years, as one of the most important inorganic membrane separation technology, ceramic membrane has been widely used in environmental protection, food, chemical, pharmaceutical and biological engineering industries. But it is unfortunately that reports about its raw material powder preparation technology are few. The size and the distribution of size of raw material powder have direct effect on support quality and reducing costs of support has been becoming the focus field in ceramic membrane support preparation recently (Qi *et al.*, 2011; Xing *et al.*, 2009), so it is very important to prepare suitable powder meet the needs of ceramic membrane support sintering with the lower cost.

Alumina powder is commonly used raw material in processes of support preparation up to now (Yang *et al.*, 2012). α -Al₂O₃ is the most stable one in a variety of the different crystal phases for its high lattice energy, high hardness, hard ground and easy agglomeration, the required grinding time is must above 12 h accord to other researchers (Chen *et al.*, 2006). So it is very important to improve production efficiency and reduce the production cost through optimizing crafts of grinding for the aim of improving technical feasibility of ceramic membrane.

Generally, sphericity of particles making by dry ball grinding method is better than making by wet ball grinding method, but dry ball grinding method is of

high energy consumption and low efficiency (Su, 2001; Zhang *et al.*, 2007; Wang and Chen, 2000). The purpose of this study is to research the best grinding condition which is probably having significant influence on the milling effect such as grinding time and the ball-to-powders ratio (Zhang and Liu, 2005) by wet ball grinding method, then evaluating the results of the experiment according to particle size and size distribution of the milled powder.

METHODOLOGY

Experimental materials: The particle size of alumina powder is of 500 meshes which were produced in a company belonging to Zibo city, Shandong province and the size distribution illustrated as follows Fig. 1.

Main instruments:

- **Laser particle size analyzer, specification:** Winner 2000S; Test range: 0.1-300 μ m; Manufacturers: Jinan Winner Particle Instruments Stock Co., Ltd.
- **Vertical circulation ball mill, specification:** LM-70; Manufacturers: Wuxi Xinbiao Powder Machinery Manufacturing Co., Ltd.

Experimental design: We set parameters as following in the wet ball grinding process, choose little ball made of α -Al₂O₃ as the milling media (Wilson *et al.*, 2011)

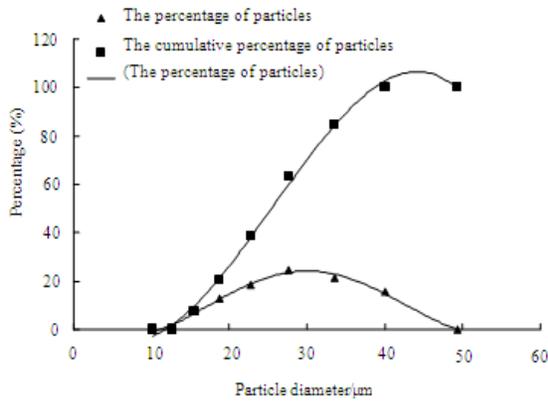


Fig. 1: Particle size and distributions of the Al₂O₃ powder

which diameter is about 6 millimeters, the ball-to-powders ratio is 3:1, 5:1, 7:1, 9:1 or 13:1, the grinding time is 2.5, 5.0, 7.5, 8.5, 10.0 or 15.0 h, respectively we will stop the experiment when the percentage of target particles having diameter less than 10 μm is higher than 90% and its size distribution is reasonable.

RESULTS AND DISCUSSION

Experimental results: The experimental result in different conditions shows as the Table 1.

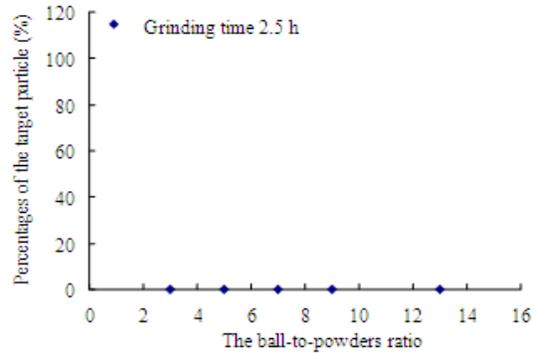
Comparison and analysis:
Influence on the milling effect in different ball-to-powders ratio conditions: In order to compare the influence more intuitive, set the percentage of the target particles as the longitudinal axis, the ball-to-powders ratio as the horizontal axis, contrast milling effect at the same grinding time and analysis the impact of the ball-to-powders ratio on the different time periods.

Figure 2a shows that the diameter of materials powder is all more than 10 μm in the initial 2.5 h on different ball-to-powders ratio conditions. Figure 2b shows that the percentage of the target particles get to 50% on the conditions of the two highest ball-to-powders ratio, but there is no target particles when the ball-to-powders ratio is lower after 5.0 h. Above shows that the ball-to-powders ratio is the most important factor in the initial 5.0 h, the higher ball-to-powders ratio the higher grinding efficiency, it is probably because rise of the ball-to-powders ratio lead to particles number generated acceleration and then lead to collision probability increased geometrically that the grinding efficiency accelerated rise or sharp rise in short time.

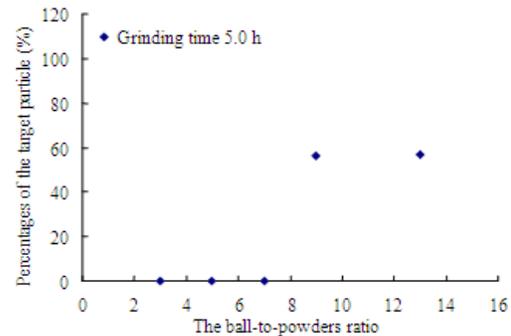
Figure 2c shows that the percentage of the target particles increase continuous and steady with the increase of the ball-to-powders ratio after 7.5 h and become into line with increase of the ball-to-powders

Table 1: Percentages of target particles in different grinding time and the ball-to-powders ratio

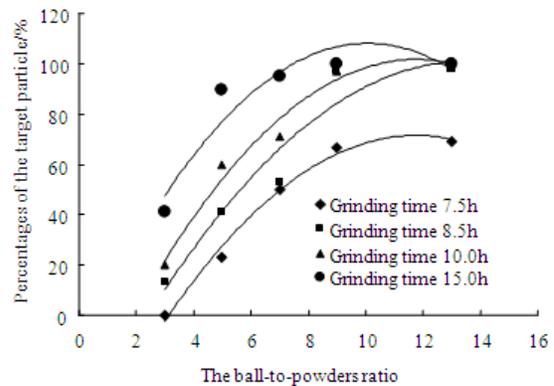
Percentages of target particles/%	The ball-to-powders ratio					
	3:1	5:1	7:1	9:1	13:1	
Grinding time	2.5 h	0.0	0.0	0.0	0.0	0.0
	5.0 h	0.0	0.0	0.0	56.0	57.1
	7.5 h	0.0	23.0	50.1	66.7	69.2
	8.5 h	13.0	41.0	53.1	97.7	97.9
	10.0 h	20.0	60.0	71.0	96.9	99.9
	15.0 h	41.2	89.8	94.9	99.9	99.9



(a) Percentages of the target particles after 2.5 h

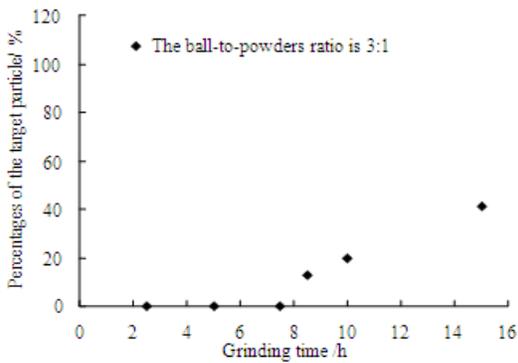


(b) Percentages of the target particles after 5.0 h

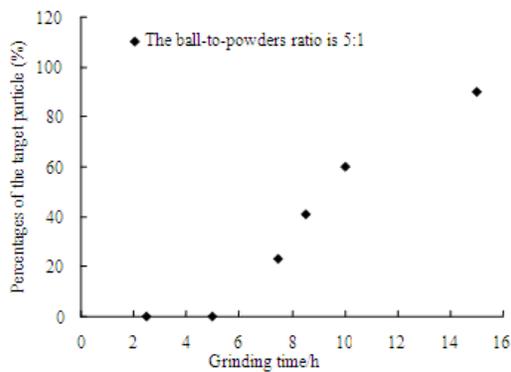


(c) Percentages of the target particles after 7.5, 8.5, 10.0 and 15.0 h

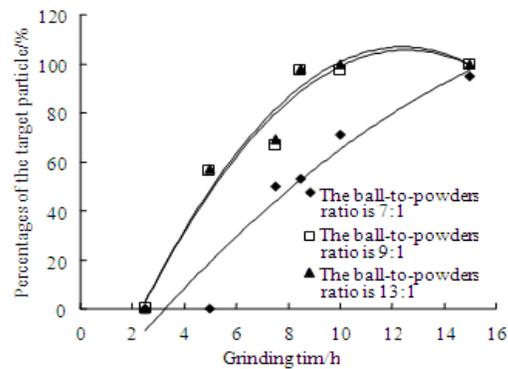
Fig. 2: Influence on the milling effect in the different ball-to-powders ratio



(a) Changes of percentages of the target particles the ball-to-powders ratio is 3:1



(b) Changes of percentages of the target particles the ball-to-powders ratio is 5:1



(c) Changes of percentages of the target particles the ball-to-powders ratio is 7:1, 9:1 and 13:1

Fig. 3: Influence on the milling effect in different grinding time

ratio when above 10:1, this shows that the ball-to-powders ratio is not the main factor on conditions of higher ball-to-powders ratio after 7.5 h, At this time, the most important factor for effective collision rising is probably the crushing of the particles themselves and the surge of the particles number.

Figure 2c also shows that only the ball-to-powders ratio greater than 9:1, grinding for 15 h or longer, it is possible to prepare suitable powder.

Influence on the milling effect in different grinding time:

In order to compare the regularity of the milling process more intuitive on conditions of different ball-to-powders ratio, set the percentage of the target particles as the longitudinal axis and the grinding time as the horizontal axis, contrast milling effect changes with grinding time on different ball-to-powders ratio conditions. The specific experimental results were shown in Fig. 3.

Overall, the collision probability between materials powder and milling media increased along with the increase of the ball-to-powders ratio and grinding efficiency increases at the same time, but the milling effect resulting trend changes inconsistent.

Figure 3a and b show that it cannot achieve experiment purpose 15 h later on the lower ball-to-powders ratio conditions. Percentage of the target particles accelerate in the all of 15-h period shows that it is a state of early crushing of materials powder and producing of small amount of target particles, the total particles number and increasing of the effective collision probability determine the whole 15-h period' grinding state, it needs more time to complete the purpose mill, of course, the energy consumption become also higher.

Figure 3c shows that the upward trend of the percentage of the target particles are no longer accelerated rise when the ball-to-powders ratio up to 7:1 and above, but the accelerate rise trend slowed down in the ball-to-powders ratio of 9:1, ball-to-powders ratio of 13:1 and it reached the mill purpose quickly when grinding time is about 8 h. At the same time, the required grinding time reduce and energy consumption reduce greatly too. On the other hand, the percentage of the target particles did not reach 100% indicating that the micron particles still have aggregation phenomena or milling media own loss produce large particles continuously (Su, 2001; Francisco *et al.*, 2009; Dai *et al.*, 2009).

Accord to Fig. 3 Basis on the final target of these series of experiments, through methods of fitting calculation, we can get the three respectively minimum grinding time is 13.7, 8.7 and 8.5 h, respectively according to three trend lines of grinding time data. This indicates under conditions of the ball-to-powders ratio is larger than 9:1, there is a little change in minimum grinding time, the ball-to-powders ratio is no longer the key factor for milling effect. In order to ensure the milling effect, combine needs of reducing energy consumption, the best ball-to-powders ratio is (9~11):1, the optimum grinding time is 9~10 h.

Table 2: The average sphere factors of particles in different grinding time and the ball-to-powders ratio

No.	Grinding time/h	The ball-to-powders ratio	Sphere factors
1	5.0	13:1	0.782
2	7.5	9:1	0.882
3	8.5	7:1	0.817
4	10.0	5:1	0.819
5	15.0	3:1	0.834

Table 3: Parameters of sintered supports

Supports No.	Porosity /%	Average pore diameter/ μm	Maximum diameter/ μm	Bending strength/ KN
1	31.7	2.1	9.0	4.5
2	35.6	2.7	9.5	4.2
3	32.7	1.9	9.5	4.0
4	34.3	2.3	9.1	3.9
5	35.1	2.3	12.0	3.9

Sintering experiments and analysis: The sphericity of the milled powder will affect the sintering process of the ceramic membrane support effect directly (Li *et al.*, 2003; Ding *et al.*, 2001), the more regular shape of the powder is, the lower porosity, the smaller contact surface among the particles and the lower strength of the sintered support have. At the same time, the particles have higher sphericity, the pore shape of the sintered supports is more regular, the flow resistance becomes lower, the support strength is more uniform, so it is necessary to study powder sphericity milled on the optimum conditions.

At first, shoot photos using an optical microscope, then scanned into a computer and then use Photoshop software, set the appropriate grid parameters and grid spacing in the photo, statistics the number of projection grid shared, contour the shared grid number of edges as well as the projection of the maximum diameter of each particle, then calculated sphere factor (Chen and Xu, 2003). The detail results were shown in Table 2. The spherical degree of the obtained powder is good; its sphere factor is between 0.78 to 0.89 and the satisfaction degree of the performance of the obtained powder to the ceramic membrane support need to confirm by excellent parameters of the support.

Sintered obtained powder milled on conditions of the ball-to-powders ratio of 9:1 and grinding time of 10 h in accordance with a certain sintering system (Ji *et al.*, 2012), we obtained 5 support samples which have reached the industrialized ceramic membrane support's parameter level. Its porosity is 31.7 to 35.6%, the maximum pore diameter is 9 to 12 μm , the bending strength is 3.9~4.5 KN, specific parameters are shown in Table 3. This shows that the obtained powder under optimum conditions meet the demand of the support well and consistent with the experimental results reported by Ding *et al.* (2008). Of course, Eliria M.J.A. Pallone has used sintered powder with high mechanical properties and hardness for the manufacturing of cutting tools (Eliria *et al.*, 2007).

CONCLUSION

The experiment shows that the best wet milling conditions is grinding time of 9~10 h and the ball-to-powders ratio of (9~11) :1. Milled particles less than 10 microns in diameter, the proportion exceeds 90% and the optimized grinding time is less than about 30% of the reported in other literatures, of course, the energy consumption is lower.

The powder sphericity is good obtained on the optimized grinding conditions and then we obtained sintered excellent support samples which have reached the industrialized ceramic membrane support's parameter level. Its porosity is 31.7 to 35.6%, the maximum pore diameter is 9 to 12 μm , the bending strength is 3.9~4.5 KN. These confirm that the obtained powder on optimum conditions meet the demand of the ceramic membrane support well.

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