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Slip cast forming of multilayer ceramic filter by fine particles migration

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Abstract

Multilayer ceramic filter was fabricated by fine particle migration occurring during the slip cast compaction of a particulate mix composed of a fine powder and coarse granules. The materials were prepared from the mixture of quartz, natural zeolite and lead borosilicate glass frit. The casting procedure involves the introduction of the powder slurry into a cylindrical shaped mold, allowing it to form a thin filtering layer onto the mold walls. The granules are then added into the mold without settlement of the previously filled slurry. The new casting method leads to a specific interlayer between the granular material and the filtering layer by a fine particle migration phenomenon. Casting, drying and sintering processes of the filters were discussed with respect to two different particle sizes of the powder slurry (δ_{50} = 0.98 μm and δ_{50} = 1.82 μm) and with variable solid concentration (5–40% solids by weight). Dilute slurry with relatively coarse size has appropriate condition for the filter fabrication.

Keywords: A. Slip casting; A. Suspensions; B. Microstructure-final; B. Porosity; D. SiO_2; E. Substrate

1. Introduction

Recently, the author has produced capillary ceramic filters using high silica ceramic powder (high SiO_2-containing glaze) by two fabrication process. The first study was similar to the conventional two layers filters; the substrate was shaped and sintered and later the filtering layer was coated by filtration [1]. The second study was a special technique in which thin filtering layers were shaped from fine particles by uniaxial pressing and later sintered at a temperature to reach for enough strength. The substrate materials were inserted between these layers and further sintering [2]. We are looking a simple fabrication process with providing high performance capillary ceramic filter with a high silica material.

It is shown that the fabrication of multilayer capillary ceramic filter is possible by fine particle migration onto granular substrate using a special casting route. The fine particle migration process was discussed in details in the filtration studies [3–9] and some approaches presented for the ceramic compaction [10–15]. The fine particles migrate through the cake pores and fill the voids at the vicinity of the bottom of the cake leading to non-uniform variations in the density across the thickness of the cake. The other consequence is the formation of a skin-layer at cake-mold interface. The high resistance layer decreased the rate of compaction and the longer casting time favours the segregation phenomena, resulting in a compaction gradient. Additionally, the deposited particles on the mold surface may be oriented and/or rearranged, and variation of local densification occurs. The non-uniform ceramic compaction also results in cracks and deformations during the drying and sintering processes.

Besides the above problems, this study is aimed to fabricate a multilayer ceramic compaction with migrating condition. If the process is successes, high performance ceramic filter has been obtained by a simple casting process and with requirement of minimum capital investment. This makes the filter wider uses in liquid/solid separation. The high silica ceramic material has low shrinkage, thus this type of materials have great potential for succession of drying and sintering without the cracking/deformation. On the other hand, it is easy to produce migrating phenomena with the silica materials.

2. Experimental

The composition for ceramic filter was designed as 86.86% SiO_2, 3.47% Al_2O_3, 5.28% PbO, 1.54% B_2O_3, 0.28% Na_2O, 0.71% MgO, 1.11% CaO and 0.11% K_2O, and prepared by

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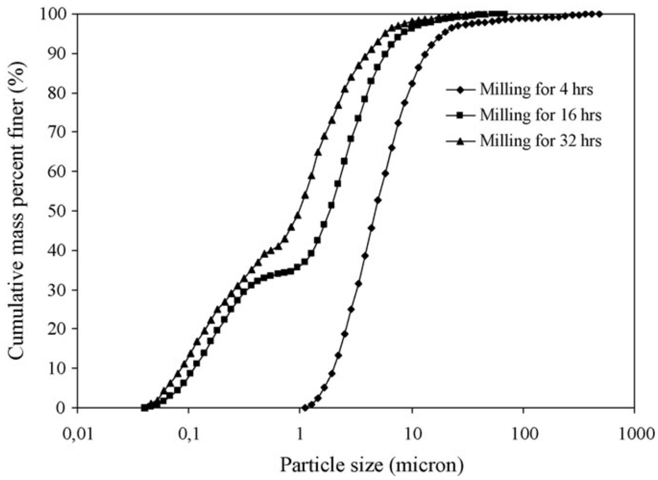

Fig. 1. Size distributions of the ceramic powders for granular core and the filtering layer.

using powder mixtures of quartz, lead borosilicate frit glass and natural zeolite (clinoptilolite). The powder mixture was ground for three different times as 4, 16 and 30 h using alumina balls in water. The particle size of the ground powders (see Fig. 1) was measured by using laser particle size analyzer (Malvern 2000). The glassy material (frit glass and zeolite) has low temperature sinterable material: the fusion point was determined by hot stage microscopy (Misura ODHT-HSM 1600/80) at about 940 °C.

The shorten time milled sample ($\delta_{50} = 6$ μm) was used for the granule production. The ground material was dried at 105 °C for 24 h, and wetted with water (5 wt.%) for agglomeration and sintered at 900 °C for 20 min with a heating rate of 5 °C/min. The agglomerates were furnace cooled. The sizes of granules are between 0.5 and 1 mm. Different solid concentrations of suspension (5, 10, 20 and 40% solids by weight) were prepared from the two fine sizes of powders ($\delta_{50} = 0.98$ μm and $\delta_{50} = 1.82$ μm). The slurries were mechanically agitated for 30 min and then casting was performed.

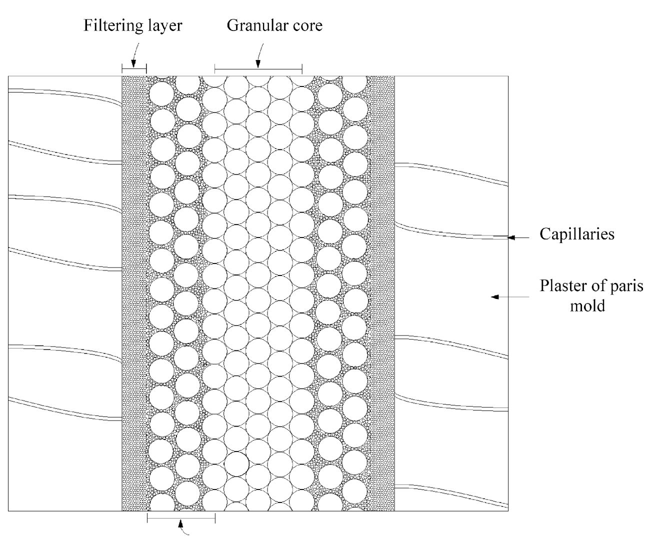

Fig. 2. Schematic representation of the casting process of a cylindrical multi-layer ceramic filter.

The important aspect in preparing the ceramic filter by means of the casting route is the multilayer compaction process. The casting unit consists of a hollow cylindrical mold, a cylindrical glass tube and a cover plate. The mold used was made of plaster of paris (produced with a plaster:water ratio of 3:2) and shaped as hallow cylinder (Ø20 mm × 100 mm). The glass tube with 20 mm internal diameter and 100 mm height tightly placed into the mold cavity. The prepared 200 ml of powder slurry was introduced into the mold with some part of slurry remained in the glass tube which is a reservoir for casting. The water of the cast slurry is absorbed into the mold walls whereupon a thin filtering layer forms on the inner surfaces. After then the granular particles were filled into the same mold in which the interior space of filter has been so filled with the granular material. The non-stop casting was achieved by the excess slurry remained in the glass tube: feeding the excess slurry into the granular medium produced a secondary compaction by the fine particle migration. By this way, an interlayer has been obtained (see Fig. 2).

The filter in the mold was allowed to dry by waiting an appropriate time such as about 30 min. Later, it was first air dried overnight in a room at 60% humidity, then, kept in ambient conditions for a day, and finally oven dried at 105 °C

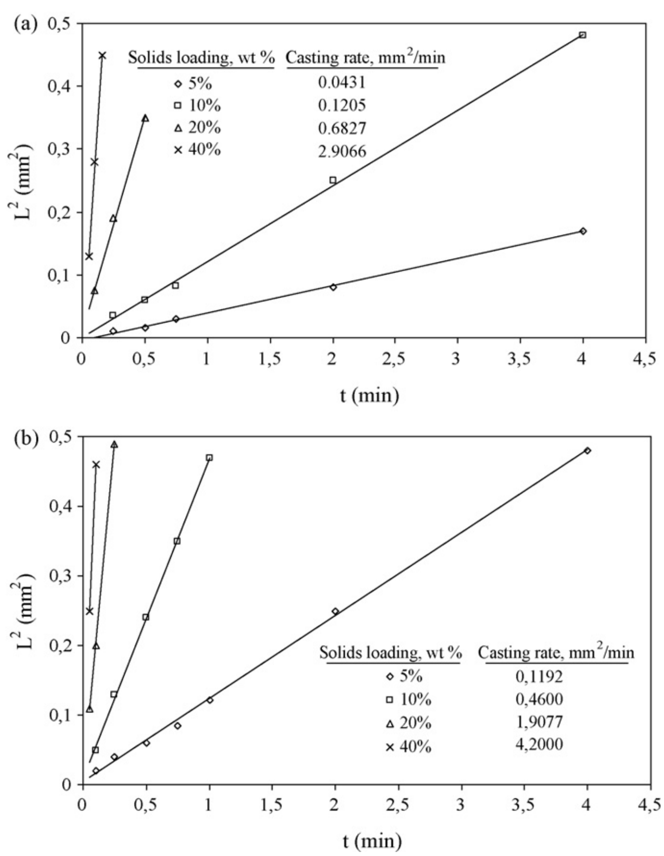

Fig. 3. (a) Parabolic dependence of the casting rates for the suspensions having relatively fine particle size ($\delta_{50} = 0.98$ μm). (b) Parabolic dependence of the casting rates for the suspensions having relatively coarse particle size ($\delta_{50} = 1.82$ μm).

for 12 h. The body was sintered at 1000 °C for 20 min with a heating rate of 5 °C/min. The microstructure of filter was investigated by scanning electron microscope (SEM) in which the fracture surface of filter was coated with a thin film of gold-palladium. The pore size range of filtering layer was determined by mercury porosimetry technique (Quantachrome Poremaster). The apparent porosity was measured by water immersion technique according to the Archimedes' principle.

3. Results and discussion

The new casting approach required for controlling of the thickness of filtering layer and interlayer between the granular-core and filtering layer (see Fig. 2). The thickness of filtering layer could be easily investigated from the conventional plot [16]: the square of cake thickness (L^2) versus casting time (t). Fig. 3a and b shows a representative plotting where L^2 versus t curves are displayed for the casting test with the powder slurry prepared with fine (δ_{50} = 0.98 μm) and coarse (δ_{50} = 1.82 μm) sizes of particles, respectively. The good linear dependence of L^2 on t is in agreement with the conventional casting rate equation. The casting rates are determined by the size of particles and solid concentration of the cast slurry in which high casting rate was obtained with the coarse particles through the dilute slurry.

A cast thickness of about 500 μm was selected for the fabrication of filters. After completion of the necessary thickness, the granular particles were filled into the interior of the cylindrical mold. After then, dead end casting was performed in which the reservoir slurries determined the

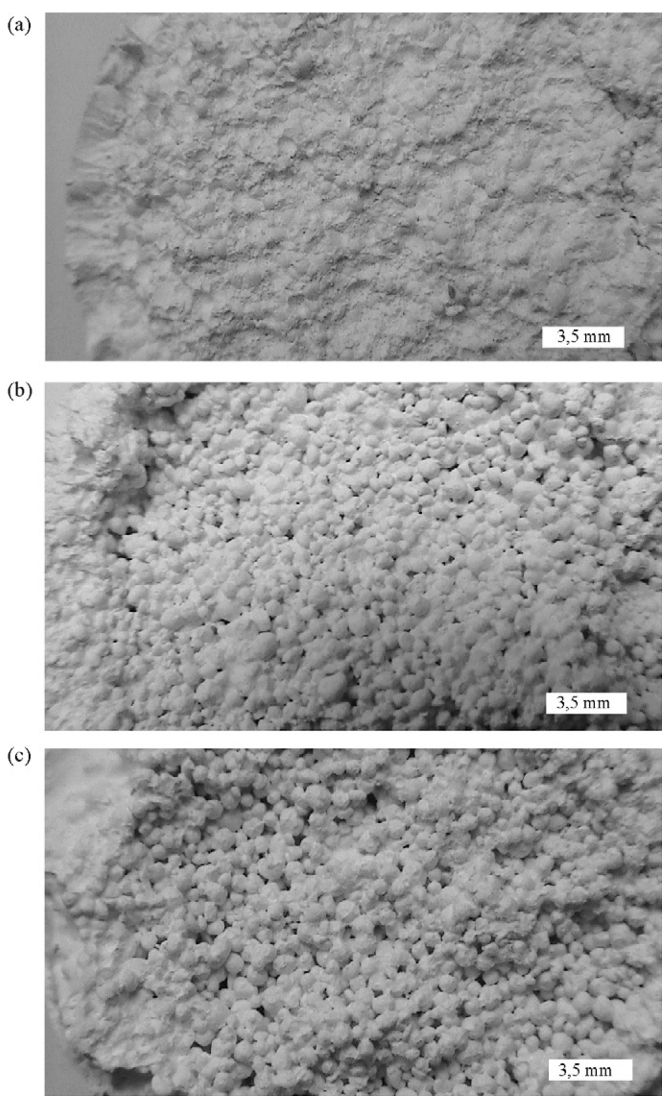

Fig. 4. (a) Photos of the granular core after casting, for the experiment conducted with the slurry containing 40% solids by weight. (b) Photos of the granular core after casting, for the experiment conducted with the slurry containing 20% solids by weight. (c) Photos of the granular core after casting, for the experiment conducted with the slurry containing 10% solids by weight.

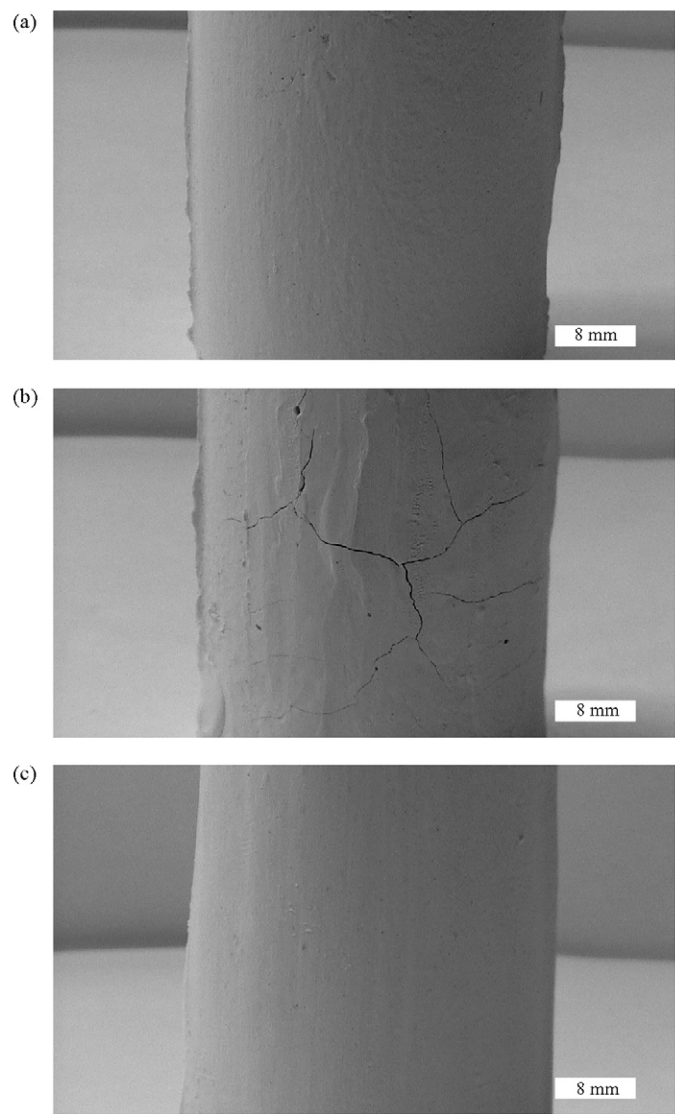

Fig. 5. (a) Photos of the surface of filter cast forming with the slurry containing 40% solids by weight with the relatively fine particles (δ_{50} = 0.98 μm). (b) Photos of the surface of filter cast forming with the slurry containing 20% solids by weight with the relatively fine particles (δ_{50} = 0.98 μm). (c) Photos of the surface of filter cast forming with the slurry containing 10% solids by weight with the relatively coarse particles (δ_{50} = 1.82 μm).

interlayer thickness. But the interlayer thickness was also determined by the occurrence of a process of particle migrating. Fig. 4a–c shows typical picture from the granular substrate after the dead end casting for the slurries 40, 20 and 10% solids by weight, respectively. It is clearly shown that a significantly thick interlayer could be obtained with the dilute slurry (10% solids by weight). The dilute slurry favours the migration of particles into pores of the granular compact. The experiments conducted with the higher concentrated slurry lead to the complete filling of pores.

It is not possible to avoid cracks on surface of the filtering layer after drying using the fine particle slurry (δ_{50} = 0.98 μm), but the higher solid content slurry gave crack free surface (see Fig. 5a and b). The non-crack surface with the higher solid slurry results from a more uniform compaction due to limited particle migration (see Fig. 4a). For all solid contents the compaction of coarse particle slurry (δ_{50} = 1.82 μm) reduces the occurrence of cracks during drying. But results are very satisfactorily with dilute slurries (see Fig. 4c). Consequently, multilayer compaction can also be obtained and these materials have potential use for the fabrication of multilayer ceramic filters. It should be also noted that casting with 5% solids takes a very long time and leads to the plaster mold saturation with water, resulting in a more complex manufacturing process.

Table 1
Thickness of interlayer with additional slurry, experiments conducted on the slurry contained 10% solids by weight

Amount of additional slurry (g)	Thickness of interlayer (mm)
No additional slurry	1
30	1.5
100	3
150	6
200	Granular substrate completely filled with the particles

It is possible to control the interlayer thickness with the dilute slurries (5 and 10% solids by weight) using additional slurries. Table 1 shows the interlayer thickness with the amount of additional slurries for the slurry contained 10% solid. The mechanical stability of filters could be increased by the thickness of the interlayer.

The filter compacted without additional slurry (see Table 1) was sintered at 1000 °C and the microstructure of filter product was investigated by scanning electron microscopy (see Fig. 6a–d). Fig. 6a shows the SEM micrograph of the fracture surface of filter which indicates the thickness of the filtering layer and the granular substrate. The granular substrate of the present filter has two layers: (i) granular core (see Fig. 6b) and (ii) interlayer

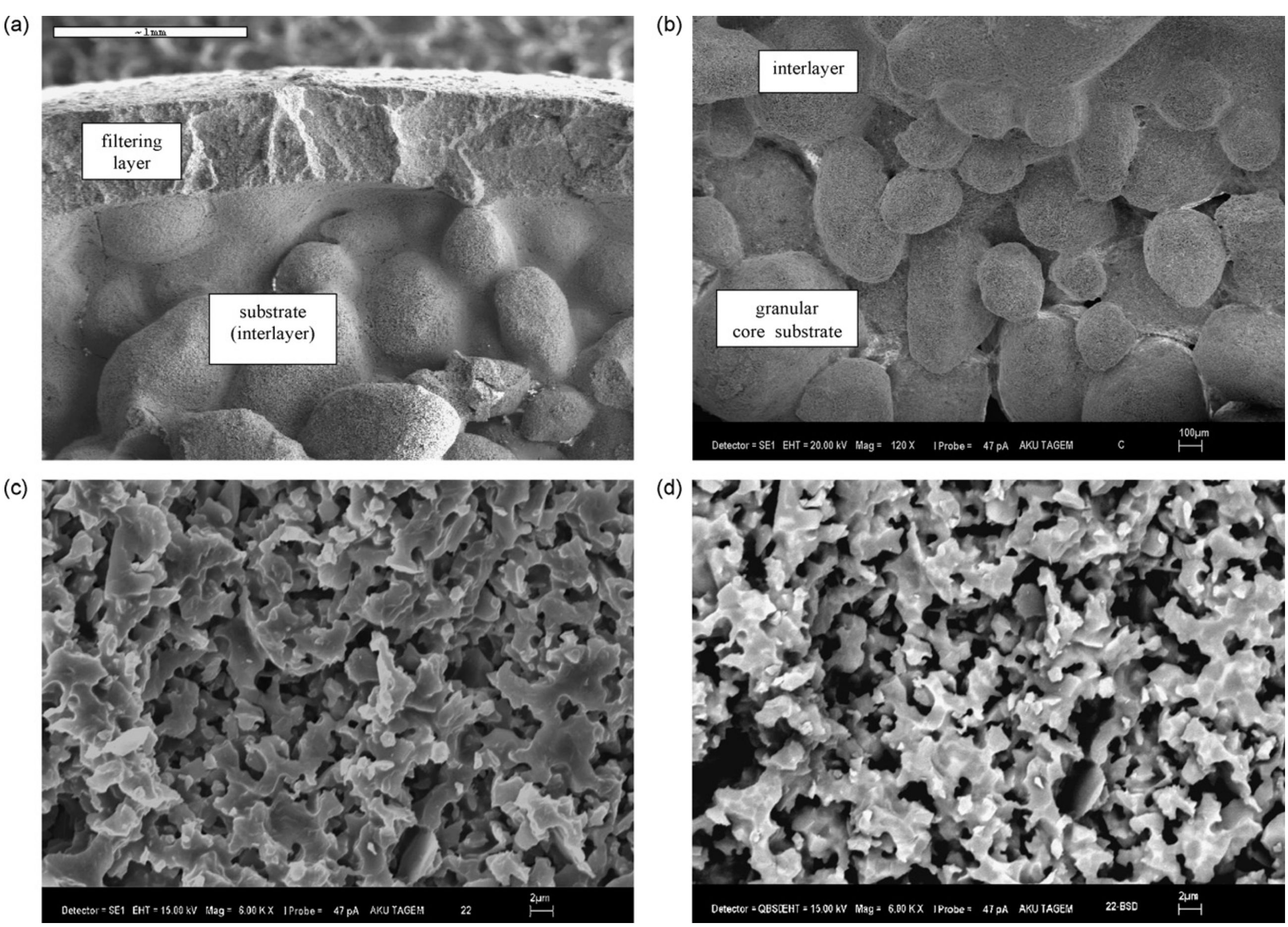

Fig. 6. (a) SEM micrograph of the fracture surface of the ceramic filter after sintering. (b) SEM micrograph of the fracture surface of granular-substrate after sintering. (c) SEM micrograph of the filtering layer after sintering. (d) SEM micrograph of the filtering layer after sintering (back scattered mode).

in which the space between the granules, along to the filtering layer, was filled by the fine particles (see Fig. 6a). Thickness of the filtering layer and the interlayer are about ~500 and ~1000 μm, respectively.

From SEM photos, it is clearly shown that the core of filters is a granular assembly were weak interconnections occurs at neck points (see Fig. 6b). It is obvious that in the core of the filter only some fine particles are observed. But the migration phenomenon of fine particles can be improved into the granular medium by favouring the liquid-flow between the granules and the settlement onto the filtering layer. By this way, the porosity of the granules assembly close to the filtering layer is low and the porosity of the core is very high. Besides, the sintering of fine particles between the granules increases the mechanical strength of the whole material.

The new cast forming route achieved the fabrication of microporous (0.3–4.5 μm) ceramic bodies with a highly porous external layer (Fig. 6c). The porosity of filtering layer was measured by Archimedes technique and found as 53.94%. At the same time the microstructure has a glassy nature (see Fig. 6d) in which the white grains with high atomic number consist of a lead containing glass homogeneously distributed through the microstructure. The glassy filtering layer makes the filter highly hydrophilic in nature. This type of microstructure with multilayer substrate is believed to have many potential applications for high performance capillary ceramic filters.

4. Conclusions

It is obvious that coating a microporous filtering layer on a substrate having large pores is very difficult. Usually, more than one layer are required for obtaining low pore size in the external filtering layer and each additional stage increase the cost of filters. Instead of the conventional application, the present approach shows the possibility of obtaining multilayer ceramic with an apparent density gradient by a simple and one stage cast-processing. Microporous filtering layer having a median pore size of 1.15 μm on to a granular assembly up to 1 mm of sizes can be obtained. The process is optimized by selecting the size of particles and the solid concentration of the cast slurry. The best filter product was obtained using relatively coarse particles (δ_{50} = 1.82 μm) and a dilute slurry (10% solids by weight).

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References

[1] O. Şan, Ç. Hoşten, Filtration testing of a ceramic capillary filter produced from a high-silica glaze, Miner. Eng. 15 (2002) 553–556.
[2] O. Şan, Microstructural characterization of capillary filter produced from a high silica-containing glaze, Mater. Lett. 57 (2003) 2189–2192.
[3] G.G. Chase, J. Arconti, The effect of filter cakes on filter medium resistance, Sep. Sci. Technol. 29 (16) (1994) 2179–2196.
[4] C. Hosten, O. San, Role of clogging phenomena in erroneous of conventional data analysis for constant pressure cake filtration, Sep. Sci. Technol. 34 (9) (1999) 1759–1772.
[5] D.J. Lee, Filter medium clogging during cake filtration, Am. Inst. Chem. Eng. J. 41 (1) (1997) 273–276.
[6] F.M. Tiller, J.R. Crump, Solid–liquid separation: an overview, Chem. Eng. Prog. (1977) 65–75.
[7] F.M. Tiller, R. Chow, Clogging phenomena in the filtration of liquefied coal, Chem. Eng. Prog. 77 (12) (1981) 61–68.
[8] M.S. Willis, I. Tosun, A rigorous cake filtration theory, Chem. Eng. Sci. 35 (1980) 2427–2438.
[9] M.S. Willis, The interpretation of nonparabolic filtration data, in: N.P. Cheremisinoff (Ed.), Encyclopedia of Fluid Mechanics, vol. 5, Gulf Publishing Co., Houston, TX, 1986, pp. 839–864.
[10] J.H.D. Hampton, S.B. Savage, R.A.L. Drew, Experimental analysis of fine-particle migration during ceramic filtration processes, J. Am. Ceram. Soc. 75 (10) (1992) 2726–2732.
[11] J.M.F. Ferreira, Role of clogging effect in the slip casting process, J. Eur. Ceram. Soc. 18 (1998) 1161–1169.
[12] P.M. Biesheuvel, Particle segregation during pressure filtration for casting formation, Chem. Eng. Sci. 55 (2000) 2595–2606.
[13] S.M. Olhero, J.M.F. Ferreira, Particle segregation phenomena occurring during the slip casting process, Ceram. Int. 28 (2002) 337–386.
[14] O. Şan, C. Özgür, The effect of fine particle migration during ceramic particle compaction by pressure casting, in: Proceedings of the 3rd International Powder Metallurgy Conference, Turkish Powder Metallurgy Association, Turkey, September 4–8, (2002), pp. 138–147.
[15] O. Şan, Some filtration parameters of diluted TiO_2/water suspensions and the cake microstructure after sintering, Ceram. Int. 32 (2006) 779–784.
[16] W.D. Kingery, H.K. Bowen, D.R. Uhlmann, Introduction to Ceramic, A Wiley-Intersecience Publication, New York, 1976, pp. 385–387.