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In situ vibration enhanced pressure slip casting of submicrometer alumina powders

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Abstract

This paper presents a novel method for improvement of particle packing in consolidation of submicrometer alumina powders by pressure slip casting. In this method, filtration cell is subjected to a mechanical vibration field with constant frequency of 50 Hz and vibration amplitudes ranging from 0 (no vibration) to 2 mm. Filtration rate, thickness and green density of the fabricated samples were measured to investigate the influence of vibration on filtration characteristics. It was revealed that employment of vibration can significantly increase filtration rate. Furthermore, there is an optimum vibration amplitude which results in the structure with the highest packing density. This value is shifted to higher vibration amplitudes as more concentrated alumina slurries is used. As the available formulation based on Darcy's law could not predict the results of the present investigation, a "*Correction Factor*" was utilized in order to increase the accuracy of the prediction in the presence of a vibration field.

Keywords: Al_2O_3; Slip casting; Shaping; Vibration

1. Introduction

Colloidal routes are the most effective methods among other consolidation techniques that offer the potential to reliably produce ceramic films and bulk forms through careful control of initial suspension structure and its evolution during fabrication. The consolidated article thus formed demands a uniform particle-packing structure for avoiding the generation of inhomogeneities that may become disproportionately pronounced in the following firing processes, such as thermal debinding and sintering.[1–3]

Pressure slip casting is one of the most widespread and accepted colloidal routes which work via fluid removal from concentrated slurries.[3,4] The green cast is formed on a porous filter which is impermeable for the particles but permeable for the liquid. Driving force for this process is a static pressure difference either is created by applying vacuum on the back-side of the filter (vacuum casting) or by applying a direct pressure to pass the liquid through the filters. Over the years, pressure filtration has been studied experimentally and modeled theoretically by numerous investigators.[3–11]

From a manufacturing point of view, a proper understanding of the influence of the relevant process parameters on the cast formation time is of crucial importance due to the increasing demand of production capacity.[10] Furthermore, a reduced cast formation time may decrease segregation effects and inhomogeneities in structure.[11–13]

However, process of filtration and expression involves a paradox: the cake formed (which is essential to cake filtration) introduces a resistance to fluid flow.[14] Because the flow resistance increases with decreasing particle size, the mechanical dewatering of fine particle suspensions may be difficult and time-consuming. To this phenomenon, a clogging of the filter medium may be added which could increase the resistance to fluid flow.[15]

Unfortunately, bridging of particles, especially when an excessive external force is applied to accelerate the settling of particles, prevents the formation of defect free and highly packed structures during consolidation of colloidal systems via pressure filtration. Besides, flocculation dramatically increases the susceptibility of particles to bridging and lead to compressible cakes which are characterized by variation of porosity in the article.[5]

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Therefore, to avoid bridging among particles and subsequent porosity buildup in the cake layer, first of all, it is necessary to satisfy the following conditions for the primary slip: (1) low viscosity and (2) good dispersion and stability. These two factors promote the particles to reach their energy-favorable positions during coalescing and formation of relatively dense incompressible cakes. In addition, the extent of the applied force determines the rate of the accumulation of particles. At high accumulation rates, the particles do not have enough time to be arranged as closed packed as possible and imperatively some voids remain among them. Deterioration of particle packing at high filtration rates has been reported in literature.[10] Furthermore, theoretical investigations of the colloidal systems which mainly focus on Darcy's law also verify this behavior.[4,6,7]

According to these previous considerations different researches were carried out to enhance filtration kinetics and simultaneously decrease the final porosity and moisture content of the residual cake.[16] Potential methods to improve the mechanical dewatering properties of conventional processes are the use of electrical,[17–19] acoustic,[20] magnetic[21] or thermal fields.[22–25]

In this work, for controlling the kinetics of process and improvement of particles rearrangement during cake formation, it has been suggested to utilize vibration during filtration process in order to help the particles to settle more effectively.

Although a process for the filtration and purification of small suspended particles in gaseous and liquid based suspensions using a high-porosity membrane situated in a resonant ultrasonic field has been reported recently,[26–28] few investigations focus on enhancement of bulk fabrication of ceramics, e.g. alumina by utilizing an external resonance to whole body of filtration cell.[20]

The effectiveness of vibration during filtration was investigated according to process parameters. As vibration has a dramatic impact on filtration rate and permeates flux, the results were adjusted with available formulation and some modifications were made to determine specific cake resistance according to vibration intensity.

2. Experimental

A commercial analytical grade of α-Al_2O_3 (Sumitomo-AKP50) with a mean particle size of 0.22 μm and a specific surface area of 11.5 m^2/g was used as starting material. Aqueous suspensions with solids loading of 5, 10 and 15 vol% were prepared using the required amount of distilled water. By addition of hydrochloric acid (HCl-37%, Reagent grade, Merck, Germany) the pH was adjusted to 4 which have been reported in many researches as the best pH for electrostatic stabilization of aqueous alumina suspensions.[29,30] Higher solids loading were avoided because of domination of van der Waals attraction on weak electrostatic repulsion and susceptibility to agglomeration due to decreasing the mean distance among colloids. Furthermore, the effects of investigating variables are less pronounced at higher concentrations.

The suspensions were milled for 2 h by an attritor using 5 mm diameter alumina balls at rotation speed of 500 rpm. The milled suspension was passed through a 180 mesh filter to minimize unintentional solid impurities and then dwelled for 30 min to settle down the un-broken agglomerates. The prepared suspensions were subjected to pressure slip casting for production of alumina samples.

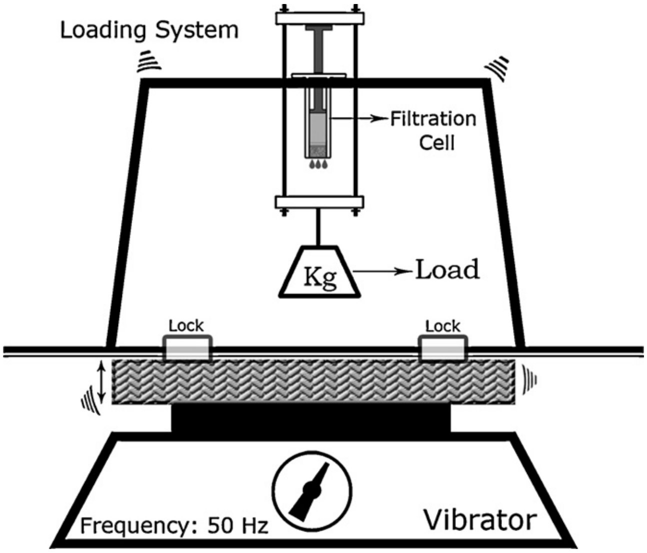

Fig. 1. Schematic of loading system and employment of vibration on filtration cell.

The filtration process was performed in small polymeric dies. A few layers of filter papers were used to separate the solid and liquid phases. 5 kg load (0.73 MPa) was applied during filtration process constantly. This load has been chosen low enough not to overshadow the influence of vibration during filtration. The initial height of the suspension in the die with respect to each solids loading was maintained at a specific level for achievement of relatively similar cake thicknesses on the membrane. The filtration apparatus including filtration cell which has been shown schematically in Fig. 1, was placed on the desk of a vibrator and a constant vibration frequency of 50 Hz with different vibration amplitude in the range of 0–2 mm where applied to the system during process.

The piston stroke, during filtration period, was recorded to determine the mean filtration rate at the applied vibration amplitudes. The green specimens was dislodged from the dies and dried in a stagnant ambient atmosphere for 24 h and subsequently pre-sintered in 800 °C for 2 h. It has been assumed that no densification occurs at this temperature and therefore the density of the pre-sintered body can be estimated as the packing properties of the cake layer formed on the filters. The density of products was measured using Archimedes principle according to the ASTM standard C20, 00.

3. Results and discussion

3.1. Influence of vibration on filtration rate

Application of in situ vibration during pressure filtration has a significant effect on filtration rate of the alumina slurries especially at lower solids content. As it can be observed in Figs. 2 and 3, during filtration process of 5 vol% alumina slur-

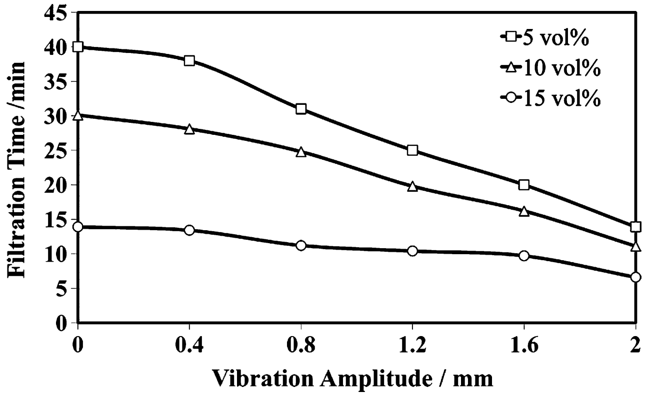

Fig. 2. Filtration time as a function of vibration amplitude at different solids content.

ries when the vibration amplitude is increased from 0 to 2 mm, the filtration time is decreased by 50% and the filtration rate is increased about two times. This increase in filtration rate can be explained as follows. External vibration field lead to oscillation of colloids in the cake around their rest point and weakening of point contacts in the cake which eventually can result in acceleration of water drainage in the pressure filtration cell. In other word, vibration can decrease the cake layer resistance to water removal.

The increase in filtration due to employment of vibration is less pronounced at higher solids content of 10 and 15 vol% because of more rapid growth of the cake layer according to slurry concentration and less water content which is supposed to pass through the filters. Moreover, influence of vibration is less perceived at relatively lower times because of rapid accumulation of particles.

3.2. *Influence of vibration on green density of alumina samples*

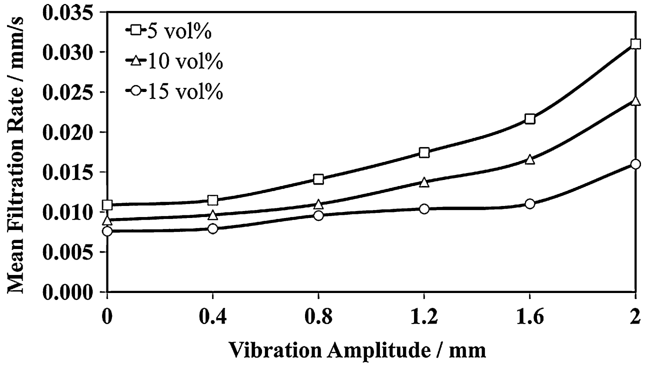

Fig. 3. Mean filtration rate as a function of vibration amplitude at different solids content.

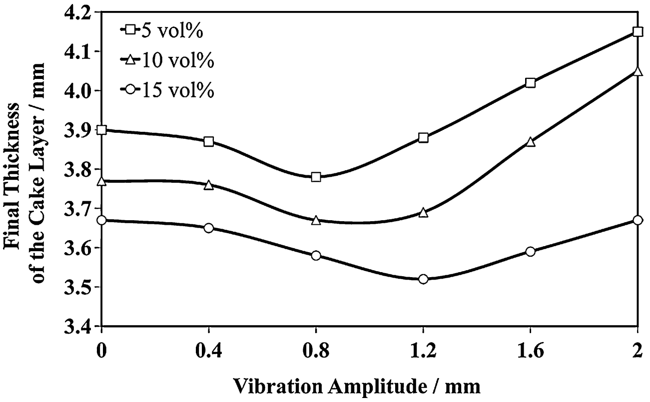

Fig. 4. Final thickness of the cake layer as a function of vibration amplitude at different solids content.

Applying vibration in pressure slip casting of alumina slurries has an enormous impact on particle rearrangement and packing during cake growth. As it can be inferred from Fig. 4 at various vibration intensities different thicknesses of the cake layer has been observed. As the solids content of all slurries are the same for each filtration process, the increase in thickness of the cake can be the consequence of porosity buildup during particles accumulation. Green density of fabricated samples which is depicted as solids line in Fig. 5 can also verify this idea about packing of structure. At relatively low vibration intensities application of vibration causes an increase in green densities of the fabricated samples in all concentrations. Moreover, according to Fig. 4, the thickness of the samples is reduced when low energy vibration is imposed to colloidal system and can be understood as an indication of better particle packing. The schematic explanation of the resultant packing of structure has been depicted in Fig. 6. This increase in packing of particles at lower vibration intensities can be demonstrated in better particle rearrangement and reduction of particle bridging during cake formation (Fig. 6B). The hydrodynamic vibration imposed on each colloid cancels out the static friction with neighbors according to drag force and put the particles in more energy favorable positions which results in denser structures. On the other hand, utilization of higher vibration intensities, detracts from the packing of structure especially at lower solids content. Relative green densities even less than the initial values (in the case of no vibration) is observed at high amplitudes of about 2 mm. Herein, increase in thickness of the green samples confirms the porous like structure of the cake. As depicted in Fig. 6C, this behavior can be originated in sever agitation and

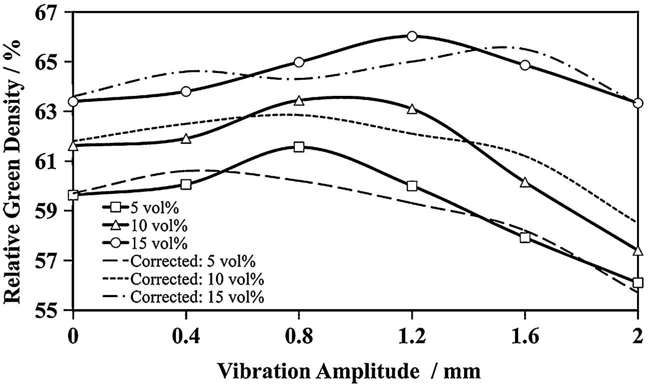

Fig. 5. Relative green density as a function of vibration amplitude at different solids content. Solids line are the actual experimental results and dashed lines are the values predicted by the modified formulation.

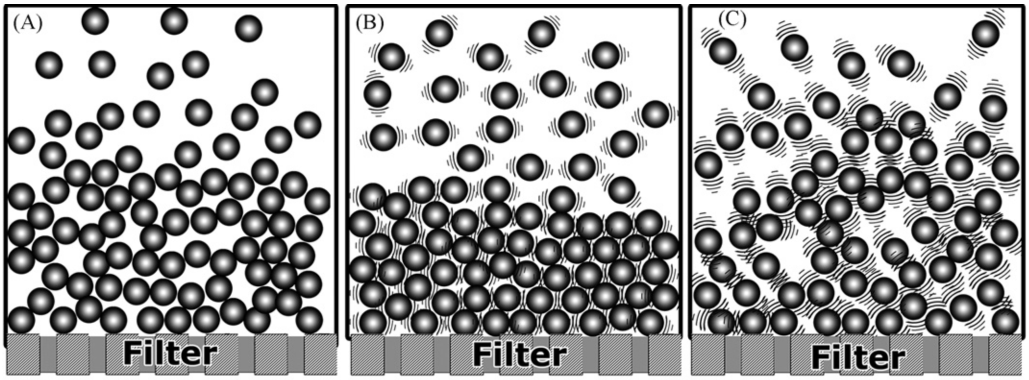

Fig. 6. Schematic of the proposed reasoning for the packing of the cake layer with respect to various vibration intensities. (A) No vibration, (B) optimum vibration intensity and (C) sever vibration intensity.

consequent bouncing of particles out of energy wells associated with high vibration energy and subsequent entrapment of large pores due to drift of other accumulating particles. Moreover, the hydrodynamic drag force accompanied with higher flow rates on the top surface of the growing cake, consume the time required for particle rearrangement and densification.

Clearly, there is an optimum vibration intensity at constant applied frequency that gives the maximum cake bed packing density.

The effect of vibration on green properties of alumina samples inevitably depends on initial solids content of the slurries. According to Fig. 5, more concentrated alumina slurries result in better particle packing. Furthermore, at lower solids loading the optimum vibration intensity is shifted to lower values in comparison with concentrated slurries in which more vibration energy is required to achieve the highest packing of the particles. The thickness of the samples (Fig. 4) also verifies the validity of this observation, which may be explained as follows. In dilute alumina slurries, at the beginning of the filtration process, the viscosity of the media is low enough for particle rearrangement. Application of excessive vibration energy to this system may inhibit the particles from being located in their energy favorable positions and can be detrimental to higher green densities. On the other hand, at relatively higher solids content, more energy is required for particle movement in relatively viscous environment. So, the effective vibration amplitude shifts to higher values as the concentration in the initial state is increased.

3.3. Modification of the existing formulation for prediction of the resultant packing of structure

Analyses of cake filtration over the past two decades have been aimed at providing a more detailed description of fluid motion through powder compacts.[7,31] Nearly all theories are based on the theory of laminar flow through a homogenous porous medium as originally introduced by Darcy in 1856.[30] Darcy's equation is normally used in filtration without sedimentation as below:

$$V_{\mathrm{W}} = \frac{\Delta P}{\mu(R_{\mathrm{m}} + R_{\mathrm{C}})} \tag{1}$$

where V_{w} is the permeate flux, ΔP is the applied pressure within the system, μ is the absolute fluid viscosity, R_{m} is the membrane resistance, and R_{C} is the cake layer resistance further defined as

$$R_{\mathrm{C}} = r_{\mathrm{C}}\delta_{\mathrm{C}} \tag{2}$$

and

$$r_{\mathrm{C}} = \frac{9\phi_{\mathrm{C}}}{2a^2}\Omega \tag{3}$$

where r_{C} is the specific cake resistance, δ_{C} is the cake layer thickness, a is the particle radius, ϕ_{C} is the cake layer volume fraction and Ω is Happel's correction factor, defined as[32]

$$\Omega = \frac{1 + (2/3)\phi_{\mathrm{C}}^{5/3}}{1 - (3/2)\phi_{\mathrm{C}}^{1/3} + (3/2)\phi_{\mathrm{C}}^{5/3} - \phi_{\mathrm{C}}^2} \tag{4}$$

of which its physical meaning is the adjustment made to obtain the true hydrodynamic force experienced by particles within a porous medium of equally sized spheres.

Combining and reconstruction of all the equations above gives:

$$\left[\frac{\phi_{\mathrm{C}} + (2/3)\phi_{\mathrm{C}}^{8/3}}{1 - (3/2)\phi_{\mathrm{C}}^{1/3} + (3/2)\phi_{\mathrm{C}}^{5/3} - \phi_{\mathrm{C}}^2}\right] = \frac{2a^2}{9\delta_{\mathrm{C}}}\left(\frac{\Delta P}{\mu V_{\mathrm{W}}} - R_{\mathrm{m}}\right) \tag{5}$$

In the case of dispersed slurries which results in an incompressible cake layer, with relatively lower porosity content and uniformity across the cake, the terms in equation five can be reasonably considered as the terminal properties of the filtration process. In other word, final packing and final thickness of the cake layer can be approximated by ϕ_{C} and δ_{C}, respectively. V_{w} can also be considered as the mean filtration rate.

Furthermore, the left side of Eq. (5) is an increasing function of ϕ_{C}. According to this equation, increasing the packing density of the cake, ϕ_{C}, results (at constant applied pressure) in a decreasing of permeate flux, V_{w}. In other word, at constant external variables, the increase in filtration rate of a slip at a fixed concentration can be the consequence of reduced cake resistivity and porosity build up in the cake layer due to inherent properties of the slurry.

According to the mean filtration rate and the final cake layer thickness presented in Figs. 3 and 4 for various vibration inten-

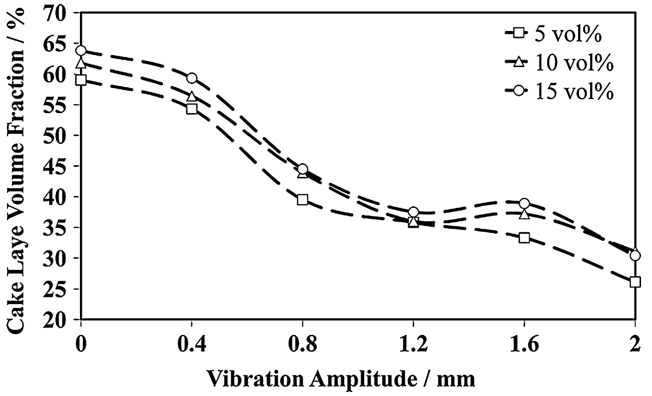

Fig. 7. Predicted theoretical volume fraction of the cake layer at different vibration intensities and solids content.

sities, it would be possible to predict the cake volume fraction by utilizing Eq. (5). A numerical solution for the above equation can give ϕ_C or cake layer volume fraction as a function of vibration amplitude which has been represented in Fig. 7. As it can be inferred when no vibration is used, the predicted values are in good agreement with the experimental results shown in Fig. 5. Nevertheless, in the presence of an external vibration field, the above formulation is failed to predict the cake layer structure. In contradiction with the theory, in this work, increasing of the filtration rate as a result of vibration intensity is not always conductive to porosity build up in the cake. As stated before, by smart selection of optimum vibration intensity the green density of the fabricated samples can improve significantly.

According to Darcy's law in Eq. (1), the only internal parameter which can be influenced by utilized vibration field is the cake layer resistance. Therefore, a modification must be employed in above formulation to predict this internal variable. The new equation for this term is suggested to follow a relation such as

$$r_C = \frac{9\phi_C}{2a^2}\Omega\Psi \tag{6}$$

where new term, Ψ, is a new term defined as "*Dynamic Correction Factor*" which can be employed when an external vibration field is applied to the filtration cell. By introducing Eq. (6) to Eq. (2) and then (1) and utilizing of actual filtration data, the dynamic correction factor evolves which is presented in Fig. 8.

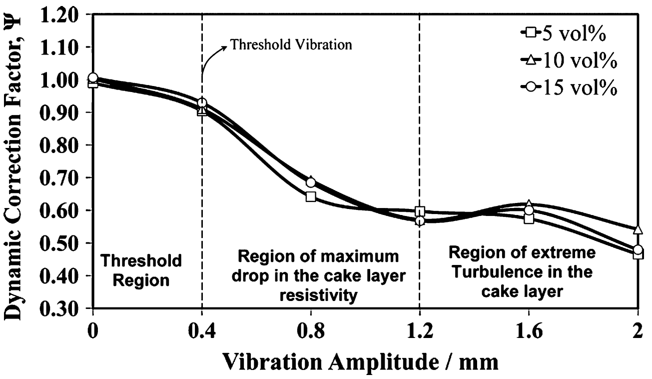

Fig. 8. "*Dynamic Correction Factor*" for modification of available formulation at different vibration amplitudes and solids content.

According to this figure, the calculated correction factor is less dependant on solids content which is a notable fact. Regression analysis of the above data and fitting of an exponential trend line, result the following relation for predicting of "Dynamic Correction Factor" in the presence of vibration:

$$\Psi = 0.978 \times \exp(0.35 \times A) \tag{7}$$

where A is the amplitude of vibration in millimeter. According to this new formulation which is independent of the solids content, the packing of the cake can be predicted more accurately probably at various solids content. Furthermore, the constant term, 0.978, can be replaced by unity (1) without much loss of accuracy. However, the results of prediction have been shown as dashed lines in Fig. 5 which are reasonably in good agreement with the actual experimental data in compare with initial prediction of formula in Fig. 7.

"Dynamic Correction Factor" was introduced to the formulation in order to modify the cake layer resistance in pressure filtration of dispersed submicrometer alumina slurries in the presence of a vibration field. According to the general form of the curves in Fig. 8 which are somehow independent of solids content, three main regions can be well defined. At lower vibration amplitudes (<0.4 mm) the effect of vibration intensity on dynamic correction factor is not very notable. In other word, a threshold of vibration intensity must be passed to influence the cake layer resistance. At medium vibration amplitudes (0.4 mm < A < 1.2 mm), dynamic correction factor is most influenced by the vibration intensity as the maximum drop is observed in this region. Probably, in this vibration intensity range, the point contacts among colloids are weakened and the carrying fluid can freely pass through the cake. At the end of this region, the turbulence of the cake layer is started which cause porosity build up in the cake layer. Very high vibration amplitudes (>1.2 mm) do not influence dynamic correction factor. It just result in sever agitation of colloids which increase the porosity of the cake layer.

4. Summary

Employment of in situ vibration during pressure slip casting can dramatically increase filtration rate and affects packing of the cake layer as a function of vibration intensity. There is an optimum vibration amplitude which results in maximum packing of structure. Application of higher intensities (more than the optimum value) can be deteriorative to the density of the article but it can increase filtration rate. The optimum value is relatively dependant on solids content which affects the viscosity. Theoretically speaking, application of vibration reduces the specific cake layer resistance but not the packing of the structure. This observation is not in agreement with previous formulations based on Darcy's law. A new dynamic correction factor, Ψ, achieved from experimental data, was introduced to modify the existing formulation of Darcy's Law in order to predict packing of the cake structure more accurately in the presence of an external vibration field.

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