# Time-Dependent Response of Coke-Glycerin Mixtures to Mechanical Vibration

**Amir Kafaei[1], Soheil Akbari[2], Simon Laliberté-Riverin[3], Louis Gosselin[4], Houshang Darvishi Alamdari[5] and Seyed Mohammad Taghavi[6]**
1. PhD Student
6. Full Professor
Department of Chemical Engineering, Université Laval, Québec, Canada
2. Postdoctoral Researcher
University of British Columbia, Vancouver, Canada
3. Postdoctoral Researcher
4. Full Professor
Department of Mechanical Engineering - Aluminium Research Centre-REGAL, Université Laval, Québec, Canada
5. Full Professor
Department of Mining, Metallurgical, and Materials Engineering - Aluminium Research Centre-REGAL, Université Laval, Québec, Canada
Corresponding author: seyed-mohammad.taghavi@gch.ulaval.ca
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**Abstract**

Carbon anodes, made by compacting anode paste in a mold through vibro-compaction, play a crucial role in the Hall-Héroult process. The response of the anode paste to mechanical vibration highly depends on the rheological characteristics of the pitch and the properties of coke particles. Given the variations in the properties of raw materials, such as coke density and shape and pitch viscosity, the dynamic behavior of the anode paste differs from time to time in the vibro-compaction stage. This variability in anode paste composition leads to different flowability and compaction capabilities; each needs a specific vibration frequency, acceleration, and time to ensure optimal compaction and rearrangement of the coke aggregates. This study aims to show how different sizes of coke particles behave regarding flowability and compaction under vibration when combined with a specific amount of a viscous fluid. To do so, we designed an experimental apparatus comprising a vibration table, a transparent vessel, and an ultra-high-speed camera to track the time-dependent motion of different materials subjected to vertical vibrations. We used a mixture of calcined petroleum coke particles and glycerin as a representative material to simulate the anode paste. Preliminary results show that the vibration rearranges the coke aggregates, minimizes the space between the particles, and compacts the bulk through time. Moreover, the particle size distribution significantly affects the compaction dynamics, with the bidisperse mixture of fine and coarse particles exhibiting the highest packing density and uniformity among the studied cases. The results are expected to apply to the industrial vibro-compaction of anode paste, and the setup can be utilized to refine vibration parameters for various simulated anode paste recipes with different flowabilities.

**Keywords:** Aluminium production, Carbon anodes, Vibration, Angle of repose, Flowability.

## 1. Introduction

The study of granular materials is essential in various industrial applications, from enhancing powder packing in pharmaceutical manufacturing to producing high-quality anodes in aluminum production [1, 2]. In particular, pre-baked carbon anodes, commonly used in the electrolysis reaction that extracts aluminum from alumina, are manufactured through vibro-compacting a mixture of 85 wt % different sizes of coke particles and butts and 15 wt % coal-tar pitch. The

pitch acts as the binder agent between the coke aggregates to cement the fine particles into the interstices of the coarse particle matrix [3].

The mixture is first poured into the vibro-compaction chamber, creating a semi-conical heap. This initial pouring stage shows inherent heterogeneities within the pile, with minimal porosity at the center and increasing porosity towards the edges [4]. In addition, the interaction of fine and coarse particles during this process leads to a preferential distribution, with finer particles accumulating in the central region and coarser ones migrating farther [5]. A descending flat plate acts as a mold and exerts pressure on the bulk of granular materials in the vibro-compaction chamber. At the same time, a vertical vibration is employed to facilitate the rearrangement of the particles. This process enables coke particles to migrate from the pile center to the sides, forming a more uniform distribution [6–8]. Given that the density and homogeneity of the produced anodes affect the overall performance of the aluminum production, an optimized vibration with specific amplitude and frequency should be applied to the system to minimize porosity variation and preferential distribution after the pouring stage. This optimized vibration setting differs from time to time, as different raw materials with varying flowabilities respond differently to vertical vibration.

The angle of repose (AoR) or slope is a key parameter that quantifies the flowability of granular materials, defined as the steepest slope of a stable conical heap formed after pouring the material onto a horizontal surface or into a confined box [1, 9–11]. The AoR is an indicator of the packing capability of a material, making it a crucial factor in many industrial applications, especially in grain storage systems where a lower AoR is desirable as it reduces the cost and complexity of filling silos and maintaining heap surfaces [12, 13].

Several factors affect the AoR and flowability of granular materials, including surface texture, sphericity, and moisture content. More textured and less spherical particles slide over each other less easily and exhibit a higher AoR due to increased interlocking and inter-particle friction [1, 5, 13]. Moreover, increasing the moisture content promotes liquid bridge formation between the particles, further increasing the AoR [14].

Additionally, particle size is another parameter that alters the AoR. Coarse particles typically exhibit a higher initial angle of repose, as they tend to interlock more and have greater friction at the contact points, reducing their flowability. Smaller particles, on the other hand, tend to flow more easily, leading to a lower angle of repose post-pouring [15–17].

In addition to the shape of the particle bed and the angle of repose, the packing characteristics of the material also play a crucial role in determining its response to mechanical vibration and final density post-vibration. The maximum packing fraction, which is the highest possible proportion of a vessel that particles can fill, is affected by the particle size distribution of a mixture. When uniform-sized spherical particles are poured into a container and vibrated, they can occupy a maximum of 64 % of the container's volume. A wider range of particle size classes, however, leads to higher packing density as smaller particles can fill the space between the larger ones, highlighting the importance of having different sizes of particles to reach the highest-packed mixture [18, 19].

In the anode production process, the paste recipe requires an optimal proportion of coarse, medium, and fine coke particles to obtain the highest anode density and mechanical strength. Sarkar et al. showed that reducing the medium-sized particles while adjusting the amount of fine and coarse aggregates can increase the green and baked anode densities and minimize the electrical resistivity. However, completely eliminating the medium-sized particles leads to a significant reduction in anode density [20]. In addition, it is shown that the particle size distribution affects the compaction behavior by determining the amount of pitch required for effective compaction. Mixtures with higher proportions of coarse particles need lower pressure to

reach the desired density. The reason is that coarse particles, having a lower surface area to volume ratio, need less pitch to coat their surfaces; therefore, more binder is left to occupy the spaces between particles, assisting their movement during compaction [21].

Previous studies in the vibro-compaction of carbon anodes have focused on the static characteristics of coke aggregates and did not utilize representative materials to resemble the behavior of the anode paste at room temperature. However, to the best of our knowledge, the time-dependent response of coke particles combined with a representative viscous fluid to vibration in the context of the anode production process has not been extensively investigated. In our recent study, we established a foundational experimental setup and methodology to assess the dynamic behavior of dry aggregates and those coated with a viscous fluid under mechanical vibrations, effectively demonstrating the differences in flowability and compaction capabilities between these two cases using an ultra-high-speed camera and a custom-built vibration setup that closely mimics the conditions of the anode production process to track the time-dependent behavior of coke-glycerin mixtures [22]. Based on our prior research, this study aims to extend the previous methodology, showing its capability to find how particle size distribution affects the compaction dynamics of anode material under vertical vibration. First, the experimental setup and the detailed procedure for pouring and vibration are described. Then, the time-dependent bulk surface is tracked during vertical vibration. Finally, the average height, height uniformity, and slope of calcined petroleum coke of different particle sizes combined with a specific amount of glycerin are compared.

## 2. Methodology

Our experimental setup, shown in Figure 1, includes a vibration table, a transparent vessel, a material feed tube, and an ultra-high-speed camera. The Vibro Pro Kinetic vibration table oscillates with a constant frequency of 60 Hz and an amplitude of 0.22 mm. Additionally, the Photron FASTCAM ultra-high-speed camera is set to capture 500 frames per second, at least eight times higher than the vibration frequency, to ensure that there are at least eight data points in each oscillation [22].

Three cases are studied in this paper: coarse particles (2.3–4.7 mm) with glycerin, mixed (50 % coarse, 50 % fine) particles with glycerin, and fine particles (0.3–0.6 mm) with glycerin. Glycerin was used as the binder agent for our experiments due to its liquid state at room temperature, which closely resembles the viscosity of the coal-tar pitch at elevated temperatures during the industrial vibro-compaction process. Specifically, glycerin exhibits a viscosity of 1470 mPa·s at 20 °C, close to that of pitch at 170 °C, 1620 mPa·s [23, 24]. This approach enables us to simulate the real industrial process without needing high-temperature handling. The total volume of material loaded into the chamber was measured in a beaker and kept at 30 $cm^3$ in all the cases.

First, a mixture of 85 % coke particles and 15 % glycerine was stirred in a vessel to ensure uniform distribution of glycerin on coke aggregates. Then, the material is transferred to the feed tube, and the sliding gate is released, allowing the material to pour gradually into a 10 cm × 10 cm × 1 cm box and form a semi-conical heap. The bounded sides of the box ensure a controlled environment for observing the compaction process, and its limited depth minimizes variations in surface profiles in the depth direction; therefore, changes in average height extracted from 2D images represent changes in the pile's overall volume. Finally, a vertical vibration (0.22 mm amplitude and 60 Hz frequency) is applied, and the bulk starts collapsing and compacting.

Following this, a comprehensive image analysis MATLAB code is developed to track the motion of the bulk coke aggregates through time. First, the captured images are cropped to a specific region of interest. The code then identifies the surface of the bulk in each image by storing the y-coordinate of pixels exceeding the preset brightness threshold. After finding the surface curvature,

the code uses linear regression to fit a line to the first half of the surface, the slope representing the angle of repose. Finally, the average height and height uniformity are measured and reported.

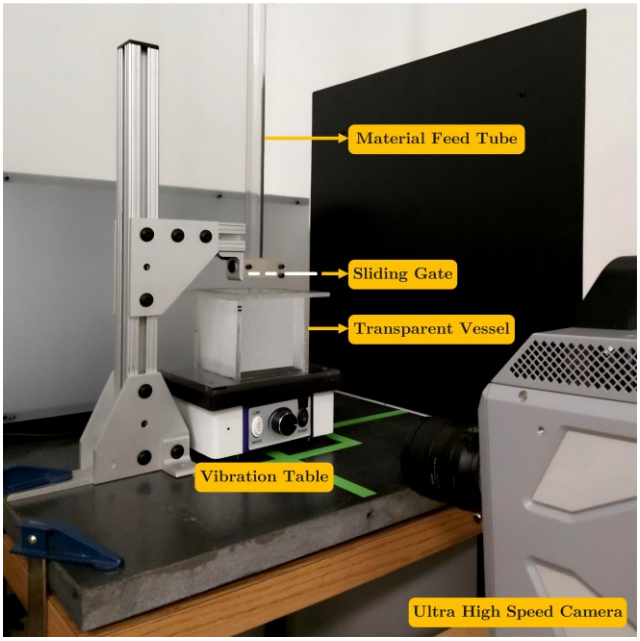


**Figure 1. Experimental setup, including a vibration table, a transparent vessel, a material feed tube, and an ultra-high-speed camera.**

## 3. Results and Discussion

Figure 2 illustrates the initial shape of the particle bed and dynamic changes in bulk surface curvature during 8-second vertical vibration for coarse particles (2.3–4.7 mm) with glycerin, mixed (50 % coarse, 50 % fine) particles with glycerin, and fine particles (0.3–0.6 mm) with glycerin. The vibration makes the heap of aggregates collapse and facilitates a more uniform distribution of particles along the horizontal axis by promoting particle-particle collisions and subsequent rearrangement of the aggregates. Additionally, figure 2 illustrates that the pile undergoes greater compaction during the initial two seconds of vibration and becomes more stable from this time onward in all cases.

The dynamic of a granular material is governed by the properties of the particles and their interactions during pouring and vibration. Among all other properties, particle size is the main factor controlling the system's behavior, interparticle friction, and packing efficiency.

Figure 3 shows the time-dependent average height of coarse (2.3–4.7 mm), mixed (50 % coarse, 50 % fine), and fine particles (0.3–0.6 mm), all with 15 % added glycerin under vibration. The average height in all cases experiences an initial dramatic fall after applying the vibration, and then it continues to decrease gradually over time, similar to the phenomena discussed in Figure 2 during the first 2 seconds. Even though the rate of decrease is less than at the beginning, there is still a negative slope after 10 s of vibration.

In addition, coarse particles show the highest overall average height after being poured into the box, as larger voids exist between them. Fine particles, on the other hand, form a heap with a lower average height compared to coarse particles. The reason is that smaller particles, with higher packing capability, form closer-packed arrangements, reducing the empty spaces between them.

(a)

(b)

(c)

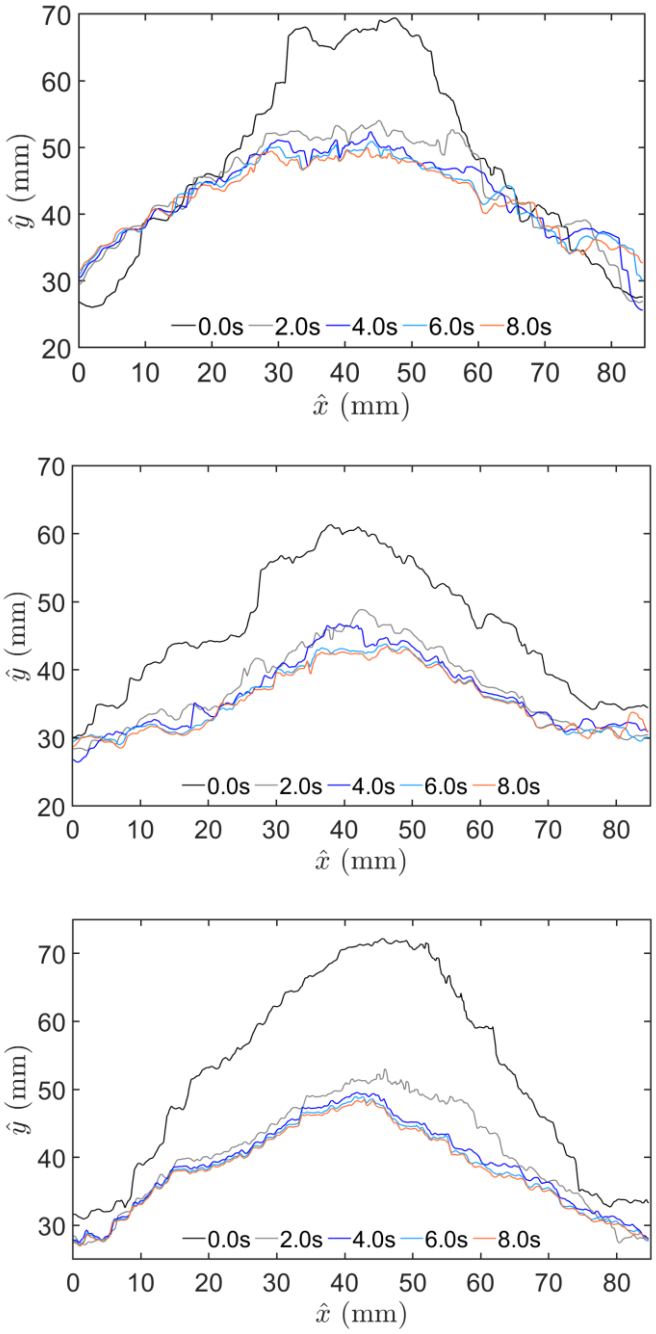


**Figure 2. Time-dependent bulk surface curvature during 8-second vertical vibration for (a) coarse particles (2.3–4.7 mm) with glycerin, (b) mixed (50 % coarse, 50 % fine) particles with glycerin, and (c) fine particles (0.3–0.6 mm) with glycerin [22].**

Finally, due to its higher maximum packing fraction, the mixture of fine and coarse particles exhibits the minimum average height during the given period. The maximum packing fraction, the highest possible proportion of a vessel that particles can fill, is a function of the particle size distribution of a mixture. The Furnas model is mainly used to estimate the maximum packing fraction of a polydisperse mixture. This method assumes that the largest particles initially fill the container with the monodisperse maximum packing fraction, $\varphi_c$. In this stage, the total volume fraction ($\varphi_{T1,Furnas}$) is as follows [25]:

$$\varphi_{T1,Furnas} = \varphi_1 = \varphi_c \tag{1}$$

Following this, the method assumes that the next smaller particle size is added to the system, filling the remaining volume fraction, $1 - \varphi_c$. The total volume fraction in the second stage ($\varphi_{T2,Furnas}$) is calculated by adding the additional volume fraction of the second particle size, $\varphi_2$, and by assuming that the monodisperse maximum packing fraction is not affected by the size of the particles for simplification [25].

$$\varphi_{T2,Furnas} = \varphi_1 + \varphi_2 = \varphi_c + (1 - \varphi_c)\varphi_c = 1 - (1 - \varphi_c)^2 \tag{2}$$

Given that $\varphi_c$ lies within the range of 0 to 1, the maximum packing fraction of a bidisperse mixture, $1 - (1 - \varphi_c)^2$, is always higher than that for a monodispersed mixture, $\varphi_c$. By generalizing this procedure for a polydisperse mixture with $n$ particle size classes, the total volume fraction is as follows [25]:

$$\varphi_{Tn,Furnas} = 1 - (1 - \varphi_c)^n \tag{3}$$

This equation shows that the maximum possible packing fraction increases by increasing the number of size classes, highlighting the importance of having different sizes of particles to reach the highest-packed mixture. This behavior is consistent with the results shown in Figure 3 for the bidisperse mixture of fine and coarse particles, in which the mixture shows the lowest average height among the three cases. In addition to the increased packing fraction for the mixture, another factor that leads to a more densely packed structure may be the fact that fine particles fill the porosities of coarse particles, enabling the particles to settle into a more compact configuration.

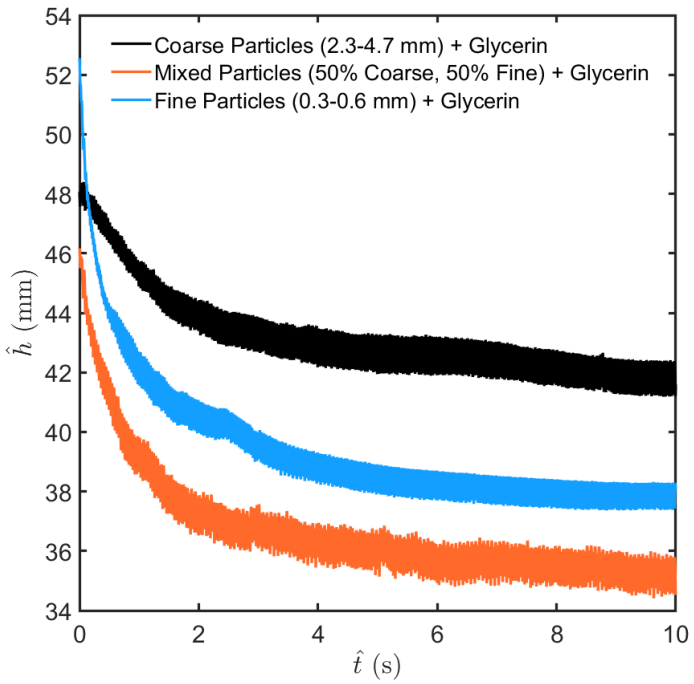


**Figure 3. Time-dependent average height during vertical vibration for coarse, fine [22], and mixed particles with glycerin.**

In addition to the average height, the uniformity of surface profiles captured in each frame is another crucial parameter. We begin by computing the standard deviation of the surface heights, which is then normalized by the range of possible y-values within the frame, resulting in normalized deviation, $\sigma_{norm}$. The height uniformity is then calculated as $1 - \sigma_{norm}$ for each frame, shown in Equation 4.

$$\widehat{hu} = 1 - \sigma_{norm} = 1 - \frac{\sqrt{\frac{1}{N}\Sigma_{i=1}^{N}(y_i - \bar{y})^2}}{Y_{max} - Y_{min}} \tag{4}$$

In which $N$ is the number of data points, $y_i$ represents each individual height value, $\bar{y}$ is the mean height of all the points, and $Y_{max} - Y_{min}$ is the range of possible y-values in the image.

A value of 1 indicates perfect uniformity, where all points on the surface are at the same height, showing no deviation among them. Conversely, a lower height uniformity indicates greater variation in surface heights, suggesting a less uniform surface profile.

Figure 4 illustrates the changes in height uniformity for particles of different sizes over a 10-second vibration period. Initially, the mixture of fine and coarse particles exhibits a more uniform surface profile, while the coarse and fine particles show roughly equal uniformity. After applying the vibration, the uniformity gradually increases in all cases as the particles rearrange and settle into a more evenly distributed pattern. By the end of the given vibration period, the mixed particles continue to exhibit the highest uniformity as the smaller particles can move into the voids created by the movement of larger ones.

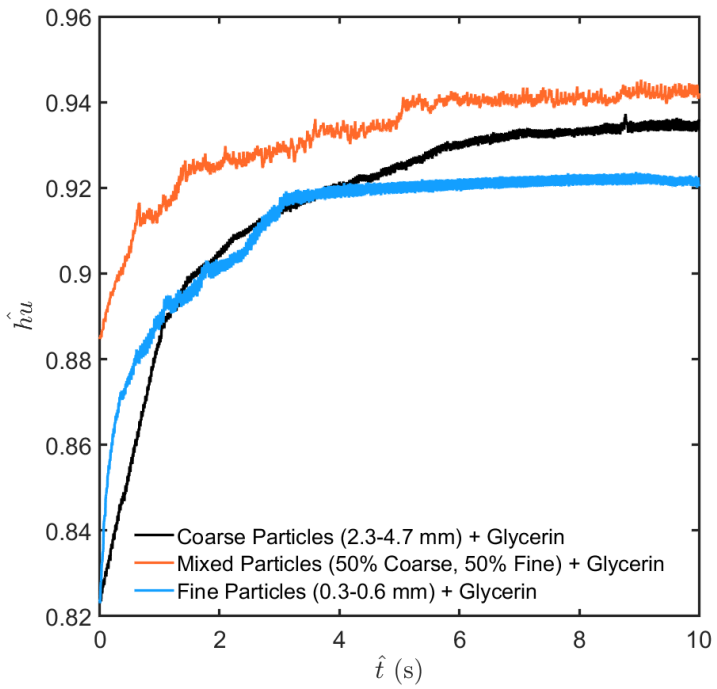


**Figure 4. Time-dependent height uniformity during vertical vibration for coarse, fine [22], and mixed particles with glycerin.**

Figure 5 shows the time-dependent angle of repose or slope for coarse (2.3–4.7 mm), mixed (50 % coarse, 50 % fine), and fine particles (0.3–0.6 mm), all with 15 % added glycerin. Analyzing the

initial angle of repose is crucial as it indicates the flowability of the material. Coarse particles show a slightly higher initial angle of repose. The reason is that larger particles tend to interlock more and have greater friction at the contact points, reducing their flowability. Smaller particles, on the other hand, tend to flow more easily, leading to a lower angle of repose post-pouring.

In addition, adding glycerin to the particles enhances viscous stresses that oppose the movement of the particles, leading to an increase in the mixture's repose angle. The effect of glycerin, however, seems to be nonuniform across different particle size classes. This may happen because fine particles experience a more significant cohesion due to a larger surface area to volume ratio, allowing them to contact the glycerin more, consequently promoting inter-particle frictions. This compensates for part of the reduction in the angle of repose resulting from their smaller size. That's why the initial angle of repose of fine particles is only slightly lower than that of coarse ones.

In addition, the angle of repose for the mixture of fine and coarse particles is initially minimal and remains the lowest throughout the vibration period. This behavior is governed by the reduced mechanical interlocking, probably because when fine and coarse particles are mixed, the fine particles fill the porosities of larger coarse particles, which mitigates the mechanical interlocking between the irregular-shaped coarse aggregates, reducing the angle of repose.

As time passes, the vibration rearranges the particles, breaks the liquid bonds between them, and promotes the flowability of the system, leading to a significant slope reduction in all cases.

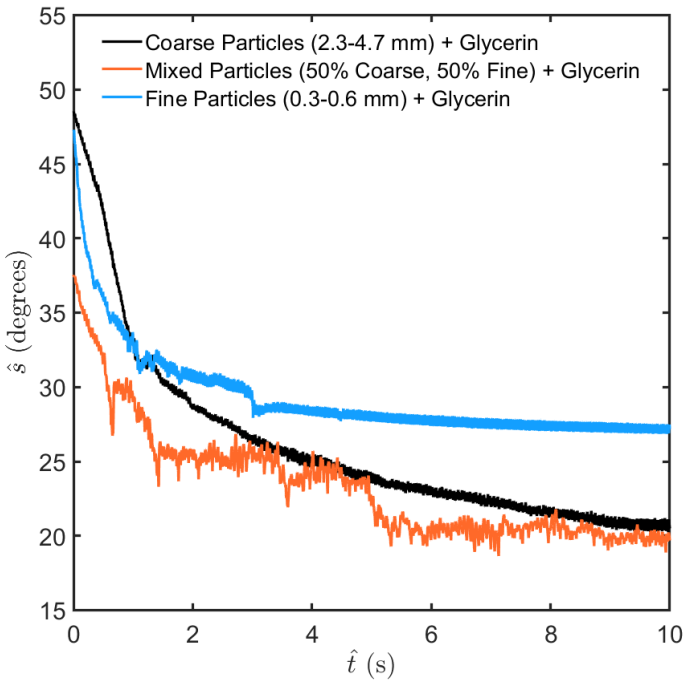


**Figure 5. Time-dependent slope during vertical vibration for coarse, fine [22], and mixed particles with glycerin.**

## 4. Conclusions

Understating the dynamic behavior of granular materials is essential in producing high-quality anodes in aluminum production, typically consisting of 15 wt % coal-tar pitch and 85 wt % coke particles. Glycerin is used as the binding agent to model the pitch since its viscosity at room

temperature is similar to that of pitch at vibro-compaction temperature. An experimental setup comprising a vibration table, a transparent vessel, a material feed tube, and an ultra-high-speed camera is used to analyze the response of different materials to vertical vibration. Three cases were studied: coarse particles (2.3–4.7 mm) with glycerin, mixed (50 % coarse, 50 % fine) particles with glycerin, and fine particles (0.3–0.6 mm) with glycerin. In general, the preliminary results are summarized as follows:

- Granular materials form a heap after being poured into a box. The vibration makes the heap collapse and compact. The results show that the particle bed compacts more at the first two seconds of vibration and becomes more stable from this time onward. This is true for both fine and coarse coke aggregates.
- The maximum possible packing fraction increases by increasing the number of particle size classes. Therefore, the bidisperse mixture of fine and coarse particles shows the highest density among the three cases over the given period of vibration.
- Glycerin acts as a binder, increasing the cohesive forces between the particles. The increased cohesiveness caused by glycerin seems to be more pronounced for fine particles, possibly due to their higher surface area to volume ratio, which increases the contact points with the glycerin and, consequently, strengthens inter-particle cohesion.
- The height uniformity of the mixed particles is initially higher than that of the other two cases and remains higher throughout the vibration.
- The angle of repose for the mixture of fine and coarse particles is initially minimal and remains the lowest throughout the ten-second vibration. This behavior can be governed by reduced mechanical interlocking, probably because when fine and coarse particles are mixed, the fine particles fill the porosities of larger coarse particles, which mitigates the mechanical interlocking between the irregular-shaped coarse aggregates, reducing the angle of repose.

## 5. Acknowledgment

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