New Insights toward the Characterization of the Carbon Paste Forming Process

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Abstract

Anode vibrocompaction plays a crucial role in defining the average density and the density gradients of the prebaked anodes. During this process, several parameters such as the frequency, the amplitude, the duration and the dead weight should be controlled. In order to improve this process, the physical mechanisms that govern the anode densification during the vibrocompaction should be understood. The aim of this work is to investigate experimentally how cyclic loading and loading rate affect the densification of a carbon paste. For this purpose, series of quasi-static cyclic and monotone dynamic compaction tests were carried out. The experimental results revealed that decreasing the amplitude of the cyclic load leads to a decrease of the stress needed to reach the target density and a decrease of the reversible component of the total deformation at the corresponding density. The results also showed that the strain rate has no effect on the compaction behavior of the carbon paste in the quasi-static regime. However, in the dynamic regime, increasing the strain rate decreases the compressibility of the carbon paste. Finally, the results obtained shall pave the way for a better understanding of the cyclic loading and the strain rate effects on the rearrangement of particles and the buildup of air pressure in the carbon paste during the forming process.

Keywords: Carbon paste, cyclic compaction tests, strain rate, air pressure, particles rearrangement.

1. Introduction

Aluminum is produced through the Hall-Héroult process where the alumina (Al_2O_3) dissolved in a bath of cyolite is reduced by a prebaked carbon anode. Carbon anodes are consumed gradually during the electrolysis process and are replaced periodically (every 25-30 days). The anode performance affects the overall energy efficiency of the electrolysis process [1]. The anode manufacturing process includes several steps that affect anode quality. First, the raw materials (calcined coke, coal tar pitch, and anode butts) are mixed at a temperature around 170 °C to form anode paste [2] . The paste is then generally formed through a vibrocompaction process at 150 °C. The resulting green anode blocks are baked in a furnace in the temperature range of 1100 °C to 1200 °C [3]. Finally, the anodes are attached to a rod with cast iron before being placed in the electrolysis cell.

The vibrocompaction process significantly affects the quality of the prebaked anodes. A poor compaction can lead to [4]:

1. High permeability, which increases the air and CO_2 reactivity of the anodes in the reduction cell;
2. High electrical resistivity (due to high porosity and fine cracks) that leads to an increase in electrical consumption and the greenhouse gas emissions [5];
3. Critical cracks in the anode that may lead to anode failure.

In order to improve the vibrocompaction process, the behavior of the green anode paste (GAP) at high temperature and under cyclic loading should be understood. Few studies have been published on the rheological behavior of the GAP [6] . The behavior of the GAP depends on the temperature of the sample and its composition [7]. Thibodeau et al. [8] performed quasi-static cyclic tests on the GAP at 150 °C in a flexible mold. The authors showed that the curvature in the strain response changes when the stress in the actual cycle reaches the maximum stress of the previous cycle. Beyond this stress, the GAP showed a softening behavior. They related this softening behavior to the breakage of aggregates that was captured by an acoustic recording system. In the same study, creep tests have been performed on the GAP. The strain response during creep stages showed a time-dependent behavior. They concluded that under static and monotonous loading, the GAP densification is dominated by the aggregates rearrangement until the solid skeleton is formed. After this point, the additional permanent deformation is resulted from the aggregates breakage. Yet, the comparison between the monotone and the cyclic compaction as well as the investigation of the cyclic amplitude effect on the densification process is not done in this work. Azari et al. [9] performed uniaxial compaction test on the GAP with different pitch/coke ratio and two quasi-static strain rates. It has been shown that the strain rate has little effect on the GAP densification. However, the effect of the dynamic strain rate on the GAP densification is not considered in this work.

To the author's knowledge, these are the only works that studied the behavior of the GAP during the compaction. There is thus a lack of information on the rheological behavior of the GAP subjected to dynamic and cyclic loadings. In this study, the behavior of an alternative carbon paste (ACP) under cyclic and high strain rate loadings is investigated experimentally. The results are analyzed in order to understand the physical mechanisms of carbon paste densification: Two hypothesis are presented.

2. Materials and Methodology

2.1. Experimental Set-Up

The experimental set-up is presented in Figure 15. A DARTEC hydraulic press is used to carry out all the compaction tests. The maximum load capacity of this press is 250 kN. The height of the sample is measured using a displacement transducer (LVDT). A thin-walled mold was fabricated for the ACP testing. The geometrical and mechanical properties of the mold are summarized in Table 10.

Table 10. Geometrical and mechanical properties of the mold.

Inner Diameter (mm)	Wall Thickness (mm)	Height (mm)	Young's Modulus (GPa)	Poisson Ratio
254	**0.356**	**140**	**220**	**0.31**

The diameter/height ratio is chosen greater than one to minimize the mold/paste friction effect on the overall compaction behavior. The mold was instrumented with four pairs of axial and radial strain gauges equally spaced on the circumference of the mold at a height of 40 mm. The strain gauges measurements are used to calculate the radial deformation of the sample during compaction using the theory of thin shells [7]. This was necessary to assure that the radial deformation is negligible and does not affect the calculation of the sample density during compaction.

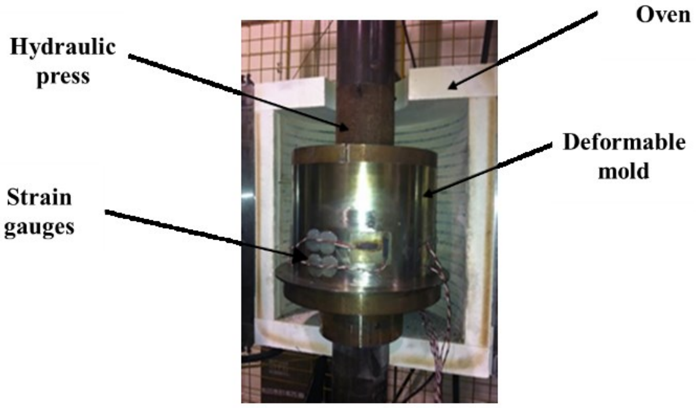

Figure 15. Flexible mold, mounted on a hydraulic press, used in all experiments.

2.2. Material

To carry out high temperature tests with the GAP, it is necessary to:

1- Control the temperature rigorously during the tests;
2- Place the anode paste as homogeneously as possible in the mold and try to start all the tests with the same GAP initial density.

However, it is extremely difficult to control the temperature during long-term tests (more than 20 min) even if the sample is placed in an oven (Figure 15). It is also difficult to put the GAP homogeneously into the mold at 150 °C since its handling is difficult. To overcome these problems and to eliminate their effects on the mechanical and thermal behavior of the samples, it was decided to work with an alternative carbon paste (ACP) at room temperature. This ACP is composed from coal tar pitch, calcined coke, and softeners. The softeners are used to have a pitch viscosity at room temperature close to that at 150 °C. The ACP is kept in barrels with hermetic closure to avoid softener evaporation that could affect the paste properties. The composition of this ACP is confidential and cannot be disclosed.

2.3. Methodology

For each test, the mold's internal wall was coated with an oil mixture to reduce its friction with the paste. Then, the mold was loosely filled with 6 kg of ACP. The initial paste height for all tests was 135 mm ± 2 mm. Two series of experiments were carried out on the ACP: (1) quasi-static cyclic compaction and (2) monotone compaction at different strain rates. Each test has been repeated two times.

2.3.1. Quasi-Static Cyclic Tests

Understanding the cyclic behavior of the ACP is very important for the vibrocompaction optimization process. Displacement controlled cyclic tests were performed and the results were compared to those of a monotonic compaction test in order to investigate the effects of cycling and cycling amplitude on the compaction. The loading path is shown in the Figure 16. The paste is compacted monotonously up to a height of 100 mm before the initiating of cycling. Each cycle consists of three stages: (1) an unloading of 8 mm, (2) a position holding (no displacement) of 10 s and (3) a loading of (8 + a) mm, where 'a' is a prescribed value. The cycling parameters were chosen in order to have: (i) a greater unloading displacement than the maximum recoverable displacement, (ii) a duration of unloading long enough to relax some of the internal stresses of the sample and (iii) a loading ‘a’ with several amplitudes. The strain rate during the tests is constant and is equal to ˙ s^{-1}. Tests were conducted for a=2 mm, a=1 mm, a=0.5 mm and a=0.25 mm. In addition, a monotone compaction test was done at the same strain rate to compare its results with the results of cyclic ones.

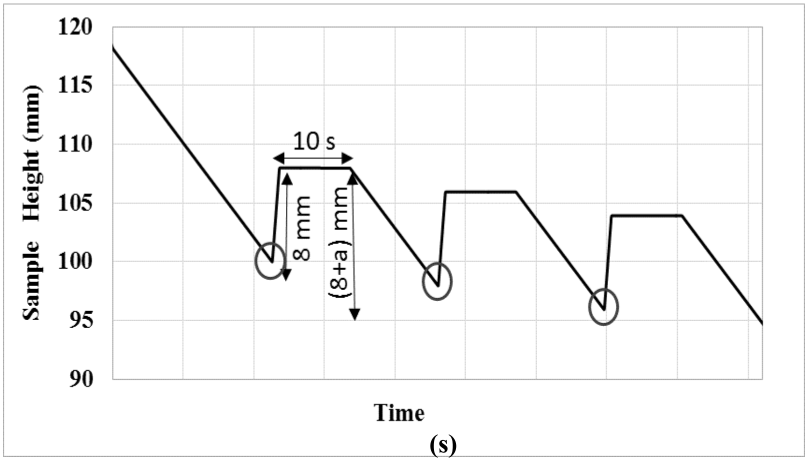

Figure 16. Loading path of quasi-static cyclic compaction.

2.3.2. Monotone Compaction Test at Different Strain Rates

Investigating the effect of the strain rate on the ACP behavior is of critical importance for the vibrocompaction application. Five monotonic compaction tests with different strain rates were performed on the ACP. The first three tests have quasi-static strain rates of: $0.0074\ \mathrm{s}^{-1}$,$0.0222\ \mathrm{s}^{-1}$ and $0.0370\ \mathrm{s}^{-1}$. The compaction in the last two tests took less than 2 seconds with the following strain rates: $0.3703\ \mathrm{s}^{-1}$ and $0.7407\ \mathrm{s}^{-1}$.

3. Results and Discussion

The mean value of the repetitions of each test are presented in the figures. The error bars represent the standard deviation (SD) to express the repeatability of the reported measurements.

3.1. Quasi-Static Cyclic Tests

Figure 17 shows the stress vs density curves for the quasi-static monotone and cyclic compaction tests with the four values of ‘a’ (Figure 16). For the cyclic tests, the points presented in Figure 17 correspond to the stress measured at the surrounded peaks of the load path shown in Figure 16.

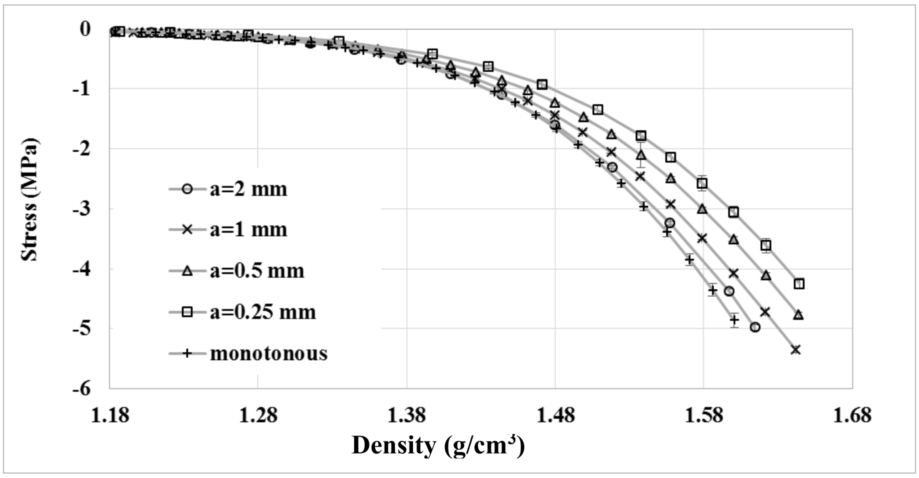

Figure 17. Stress as a function of density for the monotone and the quasi-static cyclic tests at different 'a' values.

The stress required to compact the ACP monotonously is higher than the stress required to compact it by cycling. The results showed that when the value of 'a' is decreased, the compaction requires less stress to reach the same density.

When the value of 'a' decreases, the number of cycles increases. Also, the total time of unloading considerably increases during the test. Based on the foregoing remark, the obtained results could be explained with the following two hypotheses:

HP1: The aggregates are better rearranged when the number of unloading increases. The contact forces between the aggregates decrease during unloading so that the aggregates can form a more compact configuration in the next loading (Figure 18).

HP2: The air pressure increases during compaction phases and relaxes during unloading. The decrease of air pressure during unloading leads to a decrease in the axial stress during the next loading.

A combination of both hypotheses could also be a possibility.

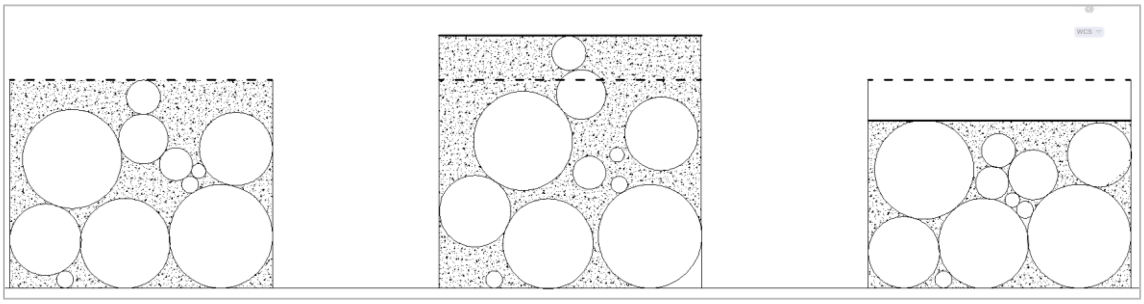

Figure 18. Rearrangement of particles during cyclic loading, from left to right: loading, unloading and next loading. Dashed line: level after the initial compaction, solid line: current phase level.

Figure 19 shows the instantaneously recoverable strain measured during the unloading phases for the four cyclic tests as a function of density. When the value of 'a' increases, the recoverable strain increases. This trend is more pronounced at the end of the tests when the density is greater.

This observation could be explained by HP2; when the air pressure decreases the recoverable deformation of the porous structure decreases, which results in a decrease of the total instant recoverable strain.

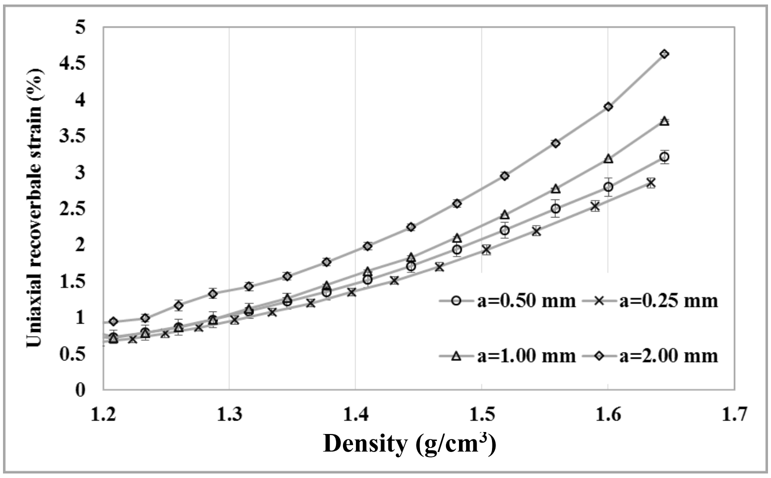

Figure 19. Instantly recoverable strain as a function of density for different 'a' values

3.2. Monotone Compaction Tests at Different Strain Rates

Figure 20 shows the stress-density curves for the five monotone tests with the following strain rates ($\dot{\varepsilon}$): 0.0074 s^{-1}, 0.0222 s^{-1}, 0.0370 s^{-1}, 0.3703 s^{-1} and 0.7407 s^{-1}. These results show that the strain rate has no effect on the compaction behavior (0.0074 s^{-1}, 0.0222 s^{-1} and 0.0370 s^{-1}) in the quasi-static regime. However, the compressibility of the sample decreases in the dynamic regime with large strain rates (0.3703 s^{-1} and 0.7407 s^{-1}). This behavior could be explained by HP2. The air does not have enough time to escape from the ACP which leads to an increase in the air pressureand the axial load.

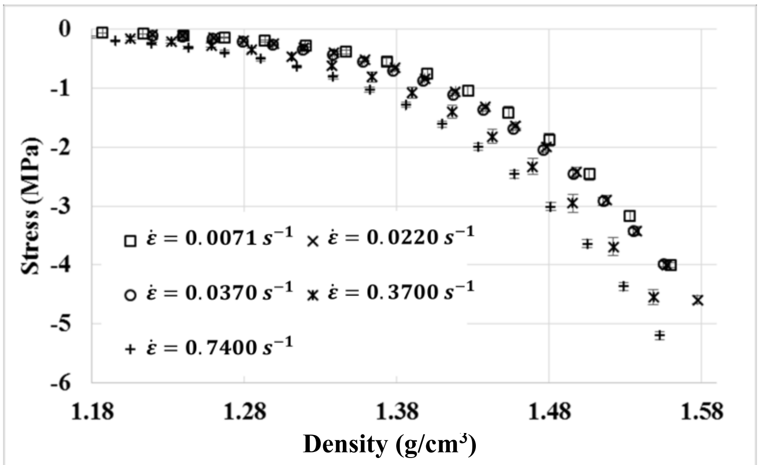

Figure 20. Effect of the strain rate on the compaction behavior.

4. Conclusions

The rheological behavior of a carbon paste was investigated experimentally at room temperature and the results are presented in this paper. Displacement-controlled cyclic tests with different amplitudes of displacement and monotone compaction tests with several strain rates were carried out. The results showed that:

1- Quasi-static cyclic compaction increases the compressibility of the carbon paste with respect to the monotone compaction.

2- Increasing the maximum amplitude of cycles increases the stress needed to reach a target density as well as the elastic deformation of the paste at a given density.
3- The strain rate of monotone compaction has no effect on the compressibility of the carbon paste in the quasi-static regime. However, increasing the strain rate in the dynamic regime decreases the compressibility of the carbon paste.

Two hypotheses were presented to explain these results. They highlight the air pressure built-up in the pores and the rearrangement of aggregates as the main driving phenomenon for the paste vibrocompaction. Further tests will be carried out to validate these hypotheses.

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