CB02 - Study of Anode Compaction Using a Model-Fluid

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Abstract

Green anodes consist of dry aggregate (coke and recycled carbon material) surrounded by a binder matrix, which is composed of fine particles and coal-tar-pitch (CTP) with viscoelastic behavior. The CTP is mixed with dry aggregate, then compacted during the forming process. In the compaction, the coarse particles rearrange while the binder matrix plastically deforms and fills in the pores with the effect of reducing the overall porosity of the green anode. Then, the green anodes are baked in a furnace where the pitch is carbonized to improve the properties of the anode. Crack formation and uneven anode density distribution are the main problems that mainly occur during the anode formation. Experimental investigation on crack formation and the development of density gradient during the compaction process is quite complicated due to several factors such as relatively high temperatures (~ 180 °C), sensitivity of pitch to the temperature variations, temperature gradient inside, and the opacity of the anode paste. We proposed a novel method to replace the binder matrix with a transparent polymer at room temperature (model-fluid) having similar rheological properties to those of the binder matrix at high temperature to perform the compaction tests at room temperature. Different polymers have been tested to tune the model-fluid to mimic the rheological properties of the binder matrix at high temperature. The real anode pastes were compacted at high temperature, and the results were compared with the compaction of model-fluid at room temperature. The results show that the compaction behavior of the new mixture (model-fluid and dry aggregate) at room temperature is similar to that of the anode paste at high temperature. This approach may allow to design further experiments in future to follow the rearrangement of coke particles during compaction and to study the effect of rheological properties of binder matrix on the paste compaction process.

Keywords: Green anode, compaction, model-fluid.

1. Introduction

Carbon anodes have a crucial role in the Hall Héroult process, and their quality directly affects the efficiency of aluminium production process [1, 2, 3]. Density gradients and cracks in the anodes are the main anode problems that occur during the forming process in which binder matrix (the mixture of the coal-tar pitch and fine particles), at relatively high temperature, moves between the coarser particles. Detailed understanding of the forming process can convey valuable information to improve the quality of the anode [4, 5]. Pitch sensitivity to the temperature variations, temperature gradient during the forming process, and dealing with opaque materials are the serious challenges in this process.

Following our previous studies [6, 7], we have proposed a novel method to overcome these challenges. The rheological properties of the binder matrix, as part of the main parameters affecting the forming process, have been obtained at high temperature [7]. Then, we experimentally studied several transparent polymer solutions at room temperature to find a transparent model-fluid that has rheological properties similar to those of the binder matrix at high temperature. In this approach, the coarser particles mix with the model-fluid to make a new anode paste at room temperature, and then the effects of operational parameters during compaction on the quality of anodes could be studied at room temperature. In the current research, we focus on finding a promising model-fluid that has rheological properties similar to those of the binder matrix at high temperature. Then, the compaction behavior of the new paste is compared with the actual anode paste during a compaction in a press.

2. Experiments

Initially, the rheological properties of the binder matrix were determined at different temperatures [7]. The results show that the binder matrix is a viscoelastic material in which the elastic and viscous properties are increased by increasing the concentration of the fine particles and decreasing the temperature.

In the experimental work, the rheological properties of an actual binder matrix were compared with those of the potential model-fluids; then, the compaction behavior of the model-paste was compared with the behavior of the actual anode paste during compaction in a press.

2.1 Rheology Tests

A Discovery Hybrid Rheometer (DHR-3), equipped with two 20 mm Peltier parallel plates, was used to characterize the rheological properties of the pitch and different binder matrices. The gap thickness was 1000 µm. The rotation and oscillation tests were performed to determine the viscous and elastic properties of the samples. Note that, in this research, we considered a binder matrix with 30 %wt. of the fine particles as a reference. The experiments show that this binder matrix is a shear-thinning material at 178 °C ($\tau = 12.6\dot{\gamma}^{0.9}$). In addition, Fig. 1 demonstrates that the viscous properties (G'') of the binder matrix are higher than its elastic properties (G') at high temperature. Therefore, we should consider two main criteria in choosing the model-fluid, shear-thinning effects with elastic modules smaller than viscous modulus.

Carbopol solution was one of the potential viscoelastic model-fluids that failed in the first step due to its plastic behavior (the results are not presented due to brevity) [8].

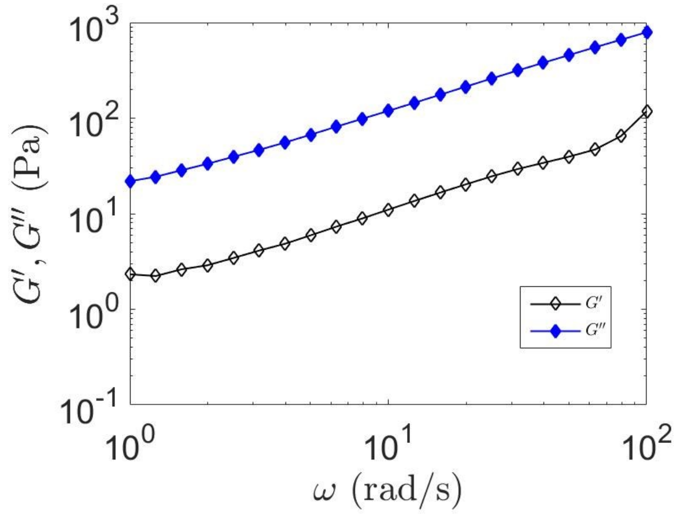

Figure 1. Elastic and viscous moduli of the binder matrix (30% of fine particles) vs angular frequency at 178 °C.

Xanthan solution was the next shear-thinning fluid (see Fig. 2). A certain amount of Xanthan powder was mixed gradually with water to prepare the Xanthan solutions. It is worth mentioning that during the preparation of samples, a small amount of glycerin was added to water to avoid any clumping. The viscosity of the Xanthan solution was increased by increasing the concentration of Xanthan which enabled tuning the model-fluid. In addition, the viscosity of the Xanthan solution drops by increasing the shear rate which is similar to the binder matrix at high temperature. Up to now, the Xanthan solution has passed the first criterion.

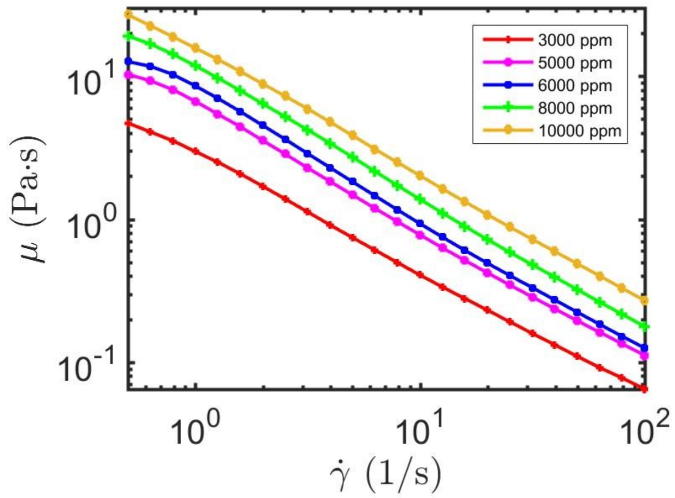

Figure 2. Viscosity of Xanthan solution at different concentrations vs shear rate at room temperature.

The oscillation tests were performed to study the elastic properties of the Xanthan solution. Fig. 3 shows that the Xanthan solution at different concentrations deliver the elastic response. However, the elastic properties of Xanthan solution are significantly higher than its viscous properties. Therefore, the rheological properties of the Xanthan solution at room temperature do not resemble those of the binder matrix at high temperature.

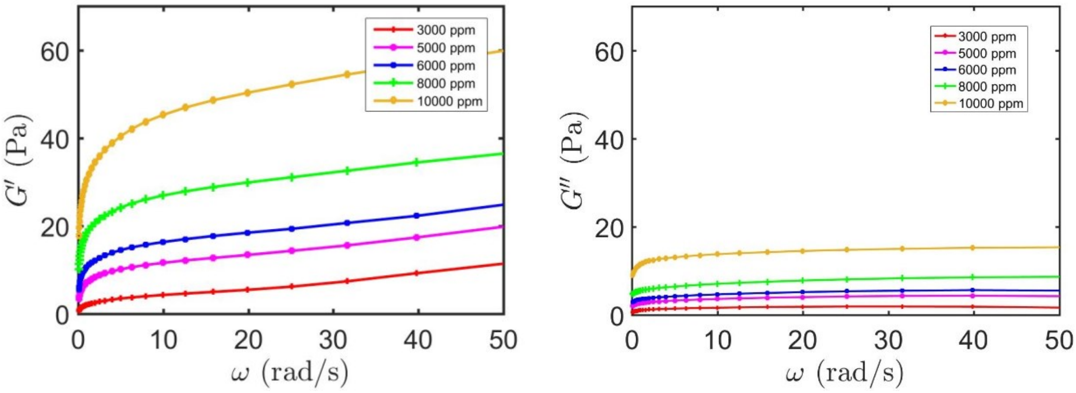

Figure 3. Elastic and viscous moduli of Xanthan solutions at different concentrations vs angular frequency at room temperature.

Polyethylene Oxide (PEO) solution is another potential shear-thinning model-fluid with elastic properties. A certain amount of PEO powder was added gradually to water and mixed for two hours at 35 °C. Then, the samples were left at room temperature for 24 h to make sure that all air bubbles were removed.

Fig. 4 presents the viscosity of the PEO solutions as a function of shear rate at room temperature. The results confirm that the PEO solution is a shear-thinning material and its viscosity increases by increasing the concentration of the polymer. Fitting the power law equation to the experimental results of the PEO solution (8 %wt.) shows that the viscosity of the PEO solution at room temperature ($\tau = 15.8\dot{\gamma}^{0.85}$) is similar to that of the binder matrix (30 % fine particles) at 178 °C.

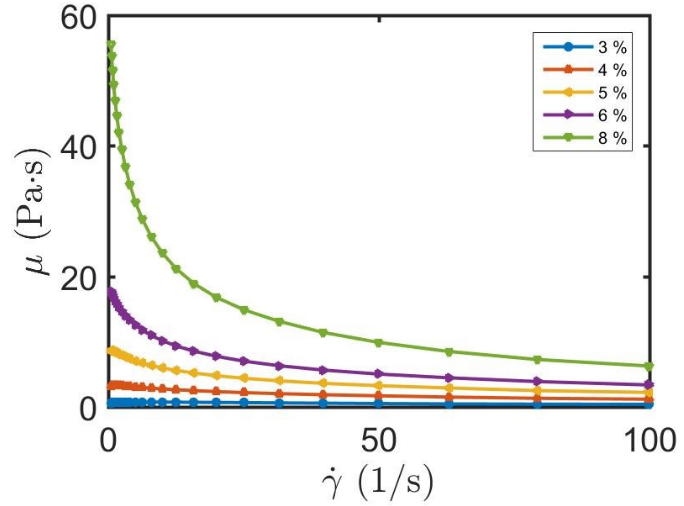

Figure 4. Viscosity of PEO solution at different concentrations vs shear rate at room temperature.

The oscillation tests of PEO solutions was the next step of the sample evaluation. Fig. 5 presents the elastic and viscous moduli of the PEO solutions at room temperature. Initially, PEO solution is a viscoelastic material and the elastic and viscous properties are increased by increasing the concentration of PEO. In addition, the viscous properties are higher than the elastic properties, which is similar to those in the binder matrix at high temperature. Therefore, the PEO solution has passed the rheological evaluations and could be a reasonable option to mimic the binder matrix in the forming process.

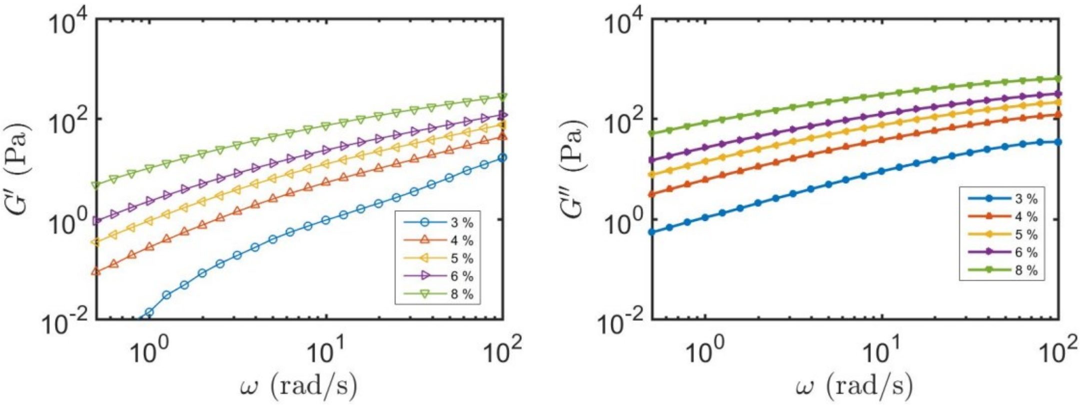

Figure 5. Elastic and viscous moduli of PEO solution at different concentrations vs angular frequency at room temperature.

2.2 Compaction Tests

The experimental compaction set-up is a DARTEC hydraulic press that is used to carry out all the compaction tests. The maximum load capacity of this press is 250 kN and the height of the sample is measured using a displacement transducer (LVDT) in a stainless-steel mold with an inner diameter of 50 mm.

Initially, an actual anode paste was prepared at 178 °C as a reference. To do so, calcined petroleum coke with a real density of 2.057 g/cm³ and a coal tar pitch with the density of 1.31 g/cm³, provided by the anode manufacturing plant, were mixed for 30 minutes at 178 °C using a Hobart N50 mixer installed in a furnace. Note that in this sample, the binder matrix contained 30 %wt. fine particles with a Blaine number of 4000 cm^2/g (11.52 g of fine particles was mixed with 26.88 g of pitch). Also, the dry aggregate was prepared using only coke particles. Further details such as the granulometric distribution of fine particles are available elsewhere [6, 7]. Table 1 shows the size distribution of the dry aggregate in the anode paste.

Table 1. Size distribution of dry aggregate.

Mesh	**4-8**	**8-14**	**14-30**	**30-50**	**50-100**	**100-200**
wt. %	0.29	0.13	0.15	0.17	0.12	0.14

The actual anode paste was placed into the mold and pressed at a loading rate of 1 kN/s with the maximum load of 140 kN at 178 °C to produce the actual green anode (AGA). The line for the green anode in Fig. 6 shows the compaction behavior of the actual anode paste (stress vs strain) at 178 °C. The initial and final heights of the sample were 80 and 50 mm, respectively.

To examine the model-fluid, sample A was prepared at room temperature by mixing the dry aggregate with the model-fluid. It is worth mentioning that in this sample, the weight fraction was respected. It means that the mass of the model-fluid in sample A was equal to the mass of the binder matrix in the actual anode paste. Comparing the results of the compaction test for Sample A (the red line in Fig. 6) and AGA shows that the behavior of this recipe is far from the actual anode paste. Incorrect proportion of the model-fluid in Sample A could be the reason for this deviation, as the density of the PEO solution was 1.012 g/cm³. To address this issue, Sample B was prepared respecting the volume fraction in the actual anode paste. To do so, the volume of the model-fluid was considered equal to that of the binder matrix, and it was mixed with the dry aggregate at room temperature. The compaction behavior of Sample B (the blue line in Fig. 6) was improved, but still did not match with that of AGA. Comparing the final heights of the compacted samples demonstrated that the final height of Sample B was greater than that of AGA (see Fig. 7). This could be related to the difference in the surface chemistry of pitch and model-fluid and greater difficulty of the model-fluid penetration into the micro pores of the particles. Therefore, we assumed that the height difference between two samples (sample B and AGA) came from the differences in the amount of pitch penetration into the pores. To prepare Sample C, we followed the recipe of sample B with less amount of model-fluid, which is equal to the final volume difference between Sample B and AGA. Note that, to keep the initial volume constant, we added the dry aggregate to the initial sample to compensate for this volume difference. The compaction behavior of Sample C was close to AGA. It means that the mixture of the model-fluid and dry aggregate at room temperature, using a modified recipe, could provide a compaction behavior similar to that of actual anode paste at high temperature.

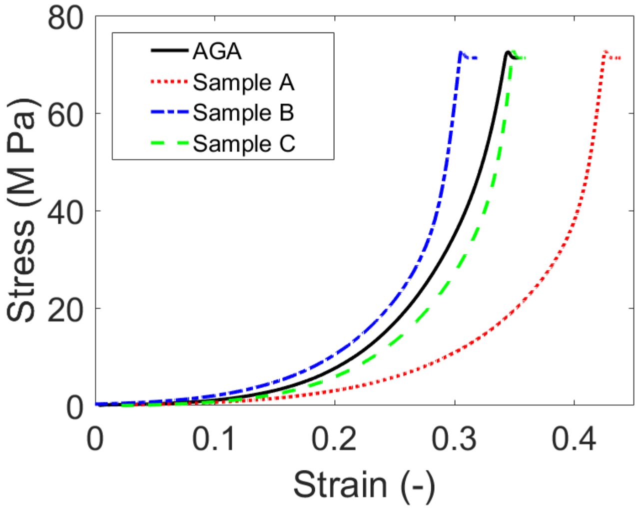

Figure 6. Monotonic compaction tests (stress vs strain) of model-green anodes and actual green anode.

Figure 7. Comparing different model-green anodes and actual green anode.

3. Conclusions

Different potential materials have been tested to find a model-fluid with rheological properties similar to those of the binder matrix. The viscous and elastic properties of the PEO solution at room temperature were close to those of the binder matrix at high temperature. The actual anode paste was prepared and compacted at high temperature as a reference. Three different samples were prepared using the model-fluid. The compaction tests confirmed that the mixture of the model-fluid and dry aggregate at room temperature, using a modified recipe, could provide a similar compaction behavior as the actual anode paste at high temperature. The following step is to use the same material in the vibro-compactor to study the effects of different parameters such as vibration frequency, amplitude, etc. on the compaction of the anode paste at room temperature.

4. Acknowledgements

This research has been carried out at Université Laval, supported financially by RÉGAL. The authors acknowledge Alcoa for the support of this work. We greatly thank Dr. H. Chaouki (Laval University) for useful discussions.

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