Rheological Characterization of Pitch and Binder Matrix

Roozbeh Mollaabbasi[1], Thierno Saidou Barry[2], Seyed Mohammad Taghavi[3], Donald Ziegler[4] and Houshang Alamdari[5]

1. Postdoc Researcher
2. Master Student
3. Assistant Professor
5. Associate Professor

Aluminum Research Centre REGAL, Laval University, Québec, Canada
4. Program Manager-Modeling
Alcoa Corporation: Aluminum Center of Excellence, Alcoa Technical Center, Pittsburgh, USA
Corresponding author: roozbeh.mollaabbasi.1@ulaval.ca

Abstract
The quality of carbon anode used in the production of aluminum is highly linked to its recipe and processing parameters. The anode compaction process is one of the critical processing steps, affecting the final density and homogeneity of the anode. A large number of parameters such as raw materials (pitch, binder and coke) properties and operational conditions affect the densification process. The combination of fine cokes and pitch results in a highly viscous material (binder matrix) which surrounds large coke particles. The binder matrix is deformed during the compaction, moving between larger coke particles, and penetrating inside their pores. Therefore, the rheological properties of the binder matrix are of primary importance in the compaction process. These rheological properties may, in turn, be affected by the binder recipe; i.e. particle size distribution of fine coke, pitch/coke ratio and processing temperature. In this work, we experimentally study the effects of mixing temperature and pitch/coke ratio on the rheological properties of the binder matrix. The data generated in this work may give a global picture of the binder matrix rheology as well as an understanding of its viscoelastic behavior.

Keywords: Rheological characterization, pitch, binder matrix.

1. Introduction

Anode is one of the most important parts in Hall Héroult process that reacts as reducing agent [1]. In order to manufacture a green anode, coke particles and pitch are blended, mixed and compacted by vibro-compactors or presses. Then, the green anodes are baked to produce the final anodes. During this process, pitch is used as a binder to fill the pores in coke aggregates and make strong coke-pitch bonds in the baked anode [2, 3]. The physical and chemical properties of coke particles, pitch and binder matrix have crucial roles in the paste quality and consequently the quality of the baked anode. Better understanding of those properties and interactions between materials during the mixing process could improve anode properties [4, 5].
The rheological properties of pitch and binder matrix are one of the main parameters that affect the quality of the baked anode [6]. Anode paste (mixture of the pitch, fine and coarse particles) is considered as a granulo-viscoelastic material. Pitch at room temperature is a solid material that behaves as a Bingham plastic at low temperature and as a Newtonian fluid in temperature between about 140 °C and 230 °C [7, 8]. The concentration of particles modifies the rheological behaviour of the paste. Hulse [9] mentioned that green anode paste has a granulo-viscoelastic behavior that depends on temperature, pitch content, coke particle size distribution, shape and roughness as well as their tendency to agglomerate.
In this article, the rheological properties of the pitch and the binder matrix at three different temperatures and four concentrations of fine particles are measured experimentally.

2. Materials and Method

2.1. Sample preparation

Calcined petroleum cokes and coal tar pitch are mixed to make the binder matrix samples that are received from the anode manufacturing plant. The density of coke and pitch are 2.057g/cm³ and 1.31g/cm³ respectively. Coarse cokes with a size range of -2.38+1.41 mm are milled in a laboratory steel ball mill for 30 minutes to produce fine particles and desired Blaine number (BN). The granulometric distribution of particles and BN are measured by a RO-TAP sieve analyser and Malvern Mastersizer 2000 respectively. Table 1 shows the size distribution and Blaine number of fine particles. A Hobart N50 mixer installed in a furnace is used to mix fine particles and the pitch in 178 °C for 10 minutes. Three samples with fine particle concentrations of 5, 10 and 15 wt. % were prepared.

Table 1. Size distribution and Blaine number of fines particles.

Size range (µm)	+150	-150+106	-106+75	-75+53	-53+38	-38	BN (cm^2/gr)
wt. %	3.1	11.5	15.6	18.7	14.6	36.5	4000

2.2. Rheological characterization

The rheological characterization of pitch and binder matrix are measured using a Discovery Hybrid Rheometer (DHR-3), equipped with two 20 mm Peltier parallel plates. The gap thickness is 1000 µm. This rheometer is able to perform the rotational and oscillational tests to measure different rheological properties of fluid such as viscosity, first normal stress difference, elastic modulus, viscous modulus etc.

3. Results and discussion

The rheological properties of the pitch and binder matrix are measured in four concentrations of fine particles and three temperatures. We use the power-law model (Equation (1)) in order to correlate the rheological behaviour of the pitch and the binder matrix (low concentrations of fine particles).

$$\tau = \kappa \dot{\gamma}^{n}, \tag{1}$$

where, τ, κ, $\dot{\gamma}$ and n are shear stress, consistency index, shear rate and power law index respectively. Table 2 shows the rheological parameters of pitch and binder matrix, considering the power law model. The pitch is a Newtonian fluid at all the studied temperatures, shown by the power index being unity. Adding the fine particles decreases the power index and the rheological properties of binder matrix deviate from Newtonian fluid. We will explain the results, including the effects of temperature and fine particle concentration more deeply below.

6. References

1. F. Chevarin et al., Active pore sizes during the CO_2 gasification of carbon anode at 960 °C. *Fuel.* Vol. 178 (2016) 93-102.
2. K. Azari et al., Influence of coke particle characteristics on the compaction properties of carbon paste material. *Powder technology.* Vol. 257 (2014) 132-140.
3. U. Suriyapraphadilok, characterization of coal and petroleum-derived binder pitches and the interaction of pitch/coke mixtures in pre-baked carbon anodes. *Ph.D. Thesis* 2008, Pennsylvania state university.
4. A. N. Adams and H. H. Schobert, Characterization of the surface properties of anode raw materials. *Light Metals* (2004), 495-498.
5. B. Madjidi et al., Simulation of vibrated bulk density of anode-grade coke particles using discrete element method. *Powder Technology.* Vol. 261 (2014) 154-160.
6. B. Majidi et al., Discrete Element Method Modeling of the Rheological Properties of Coke/Pitch Mixtures,. *Materials.* Vol. 9 (2016) 334.
7. S. Hlatshwayo et al., Rheological behavior and thermal properties of pitch/poly(vinyl chloride) blends. *Carbon*. Vol. 51 (2013) 64-71.
8. St. Thibodeau et al., New insight on the restructuring and breakage of particles during unaxial confined compression tests on aggregates of petroleum coke. *Powder tehnology.* Vol. 253 (2014) 757-768.
9. K.L. Hulse, Rheological behavior, Anode manufacture: Raw materials, formulation and processing parameters, (2000) 283–334.
10. E. Guth, Theory of filler reinforcement. *Journal of Applied Physics*. Vol. 16 (1945) 20.
11. T. G. Mezger, The rheology handbook, 2nd eddition (2006).
12. A. Eslami and S. M. Taghavi, Viscous fingering regimes in elasto-visco-plastic fluids. *Journal of Non-Newtonian Fluid Mechanics*. Vol. 243 (2017) 79–94.