Powder Technology 254 (2014) 228–234

Contents lists available at ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

Analysis on the die compaction of anode paste material used in aluminum production plants

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ARTICLE INFO

Article history:
Received 8 August 2013
Received in revised form 7 January 2014
Accepted 8 January 2014
Available online 16 January 2014

Keywords:
Anode paste
Coke
Die compaction
Pressure
Pitch
Relative density

ABSTRACT

The quality of anode electrodes is an important issue in aluminum production plants. Therefore, not only raw material conditions but also the conditions of different production steps could affect the anode quality. Analyzed in this paper is the die compaction of some anode paste samples with specified compositions. Portions of fine coke and coarse coke particles were mixed and some coal tar pitch was added to this mixture in order to make the anode paste sample. The Blaine number of the fine coke portion and the pitch content were selected as two variables to make different anode paste samples. Thereafter, each sample was compacted in a rigid die while applying a uniaxial stress up to 60 MPa. The compaction behavior of different samples was studied and an analytical relationship between the green relative density and the axial pressure applied in the die was suggested. After having identified the parameters of the suggested expression for each sample, the contribution of the rearrangement phenomenon to the material densification was studied. Regarding the good agreement between the suggested relationship and the experimental results, it would be possible to predict the compactability of different anode pastes based on the analytical equation.

1. Introduction

In aluminum production plants using Hall–Héroult electrolysis process, the following anodic reaction occurs:

$$C + 2O^{2-} \rightarrow CO_2 + 4e. \quad (1)$$

As seen by reaction (1), the anodic reaction leads to gradual consumption of carbon that is the main element of anode electrode. However, some undesirable reactions may happen that lead to the overconsumption of electrode:

$$C + CO_2 \rightarrow 2CO \quad (2)$$

$$C + O_2(\text{air}) \rightarrow CO_2. \quad (3)$$

These undesirable reactions are usually called as the gasification reactions. The electrode overconsumption is a key factor influencing economical aspects of aluminum production. The paste used for the fabrication of anode electrodes consists of fine and coarse coke particles as well as coal tar pitch. In technical terminology, the mixture of fine coke and pitch is called 'binder matrix' and the coarse particles are referred to as 'aggregate'. A major issue is to have a material structure that ensures not only an appropriate electrical conductivity but also a minimal amount of carbon gasification. Material density and its distribution throughout the electrode (or equivalently the porosity and pore structure) can be considered as important properties that could affect the function of electrode. It is evident that material density is influenced by both the materials' inherent properties and the anode fabrication conditions. The fabrication process generally consists of material preparation, mixing different constituents that will form the anode paste, compaction of the anode paste to the desired shape to obtain the green compact; and finally baking of the compact in order to obtain the final product with the desired characteristics. The effect of different parameters on the resulted structure of anode material has been the subject of many researches. For instance, in a study conducted by Hulse [1], the structure and subsequently the properties of anode were found to be principally related to pitch content and size distribution of coke particles. Smaller particles can fill the space between the larger particles [2,3]. Thus, size distribution of coke aggregates as well as size distribution of fine coke affects the maximum density [2], pore size distribution [4] and electrical resistivity [5] of anodes. Shape, roughness and porosity of coke aggregates also influence particle flow, packing and consequently the anode density [2,6,7].

The influence of fineness of fine coke has been studied in some other works [8–11]. Fineness is determined by Blaine number (BN) given in cm^2/g that is obtained by Malvern analyzer based on a permeability experiment. It should be pointed out that BN may differ from the

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http://dx.doi.org/10.1016/j.powtec.2014.01.033

Table 1
Particle size distribution, Blaine number of fine coke.

Blaine number (cm^2/g)	Particle size distribution (wt.%)				
	+ 149 μm	− 149 + 74 μm	− 74 + 53 μm	− 53 + 37 μm	− 37 μm
2300	22.8	35.5	10.7	10	21
4000	1.6	26.4	20.2	16	35.8
6300	0	10.7	20.6	26.2	42.5

exact value of specific surface area. This number is usually employed as a comparative variable.

In addition to the granulometry of the coke, pitch content is an effective factor for anode quality. McHenry [12] and Hulse [1] showed that anode characteristics such as baked density, electrical conductivity and compressive strength improve with increasing pitch content to an optimum amount. Optimum pitch content in the paste that wets the particle surface and fills the interparticle spaces depends on granulometry and surface properties of raw materials.

Binder matrix, globally known as a viscous material, fills the interparticle spaces in the anode paste. Previous studies show that the fine coke content influences binder matrix viscosity [13–15]. When fine coke exceeds 50 wt.%, the behavior of binder matrix changes from Newtonian to viscoelastic [16,17]. For viscoelastic materials, both viscosity and elastic behaviors contribute to deformation and compaction. The viscosity of binder matrix decreases with increasing temperature, pitch content and particle size [13,14,18,19] and thus the contribution of viscosity to the compaction becomes significant [15]. This means that the compaction behavior of the material becomes more sensitive to loading rate.

Anode making literature is principally focused on the influence of pitch content, granulometry and amount of fine coke on filling the voids between the coke aggregates. Apart from a few works on some similar materials such as the one of Gethin et al. [20] carried out on the compaction of carbon powder, there is a serious lack of information about the effect of paste parameters on its compaction behavior. In this work, different anode samples with different pitch contents and Blaine numbers were compacted in a rigid die. Then, from a phenomenological point of view, the compaction behavior of samples and the evolution of density were studied and compared.

2. Materials and experimental procedure

A calcined petroleum coke with a real density of 2.057 g/cm^3 milled in a laboratory-scale ball mill and fine coke particles with three different granulometries were produced. Blaine number of the fine coke fractions was measured by a laser diffraction particle size analyzer (Malvern Mastersizer 2000) and the size distribution was evaluated by sieve analysis. Table 1 shows the size distribution of three groups of fine coke particles used in this work with Blain numbers of 2300, 4000 and 6300 cm^2/g. It should be pointed out that the Blaine numbers were not presented in SI unit (m^2/kg) in this work because Blain number is usually reported in c.g.s unit system in technical texts. The prepared fine coke and coarse coke particles were mixed with the required amount of pitch to obtain anode paste samples with different weight ratios of pitch to coke (P/C ratio). The weight fractions of fine coke and coarse particles are cited in Table 2. Mixing was performed at 178 °C for 10 min in a Hobart N50 mixer that was installed in an oven. It should be noted that the pitch had a Mettler softening point of 109 °C and a quinoline insoluble (QI) content of 15.5%. The total mass of coke and pitch was 488 g for each sample.

The paste samples were compacted in a cylindrical rigid die with an inner diameter of 68.3 mm (Fig. 1a). The die was placed inside a tube

Table 2
Granulometry of coke particles in the paste samples.

Size range (US mesh)	− 4 + 8	− 8 + 16	− 16 + 30	− 30 + 50	− 50 + 100	− 100 + 200	Fine coke
wt.%	22.0	10.0	11.5	12.7	8.8	10.8	24.2

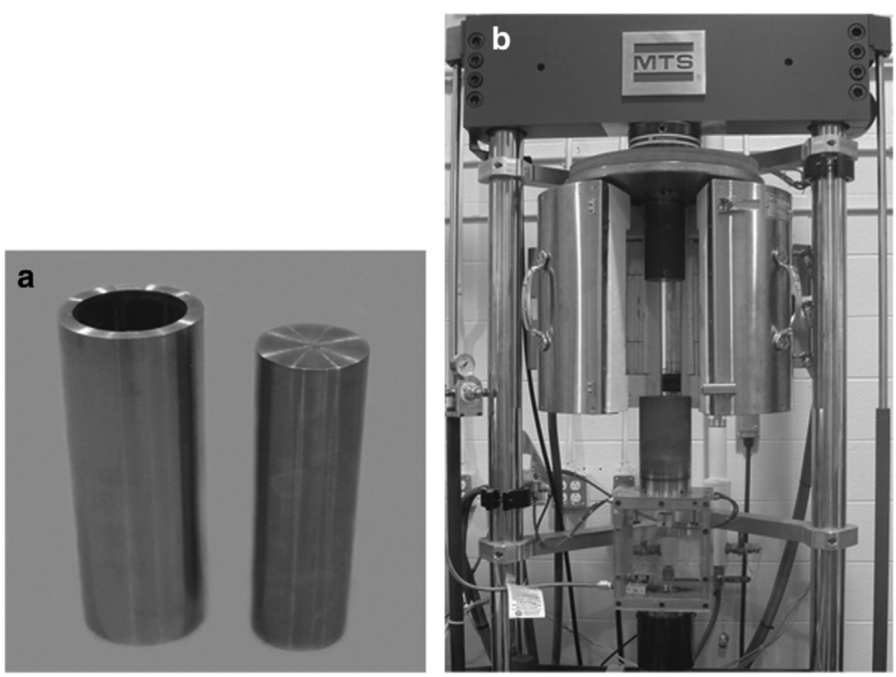

Fig. 1. Equipment used for compaction tests. (a) Die and punch, (b) servohydraulic press.