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Influence of coke particle characteristics on the compaction properties of carbon paste material

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ABSTRACT

The anode electrodes in electrolytic production of aluminum are composed of coke particles and binder pitch. A paste consisting of the particles and pitch is made and compacted to form the green anode. The influence of coke particle properties including shape factors and density on the particle packing, compaction behavior of the paste and the final density of green anode was studied. Five sponge cokes and one shot coke from different industrial sources were used in this study. The shape factors and apparent density for different size fractions of particles were measured and the volume fraction of open and closed pores within the particles was determined. The bulk density of the particle bed was measured with and without applying vibration. Inter-particle void fraction was then calculated to determine the packing behavior of the particles. The pastes were made and compacted to make laboratory-scale anodes. It has been revealed that particle shape is an important factor determining the packing properties and compacted density.

1. Introduction

Carbon anodes are used in electrolytic production of aluminum to transfer the electric current to the electrolysis bath. Calcined petroleum coke is mixed with coal tar pitch (binder) to make a carbonaceous paste. This paste is compacted through vibro-compacting or in some plants by pressing. Coke aggregates rearrange in the binder media during the compaction process to reduce the voids. The green anode is then baked to achieve the required strength and to improve electrical conductivity through carbonization of the binder. Making homogeneous anodes with high density is of great interest in order to decrease electrical resistivity and to increase energy efficiency in aluminum smelting process. There is a relationship between the green anode characteristics and final properties of baked anode. Density of green anode determines the final density, electrical conductivity [1] and mechanical properties [2] of the baked anode. Therefore, studying the green anode density gives an indication on the quality of baked anode. Density of green anode depends on the compaction behavior of the paste which in turn depends on the paste formulation and anode making parameters. This work aimed at studying the influence of density and shape factors of coke particles on the compaction behavior of anode paste.

Size distribution and internal porosity of coke particles are not the only factors affecting the bulk density [3]. Shape and roughness of coke aggregates have an influence on particle flow and packing, and consequently on the anode density. Coke particles with more irregular shapes and higher roughness have higher inter-particle friction and inferior flowing and packing properties [3,4]. Therefore, cokes with lower internal porosity will not necessarily result in denser anodes, instead, both internal porosity and shape of particles will influence the anode density. Belitskus [5] confirmed that spherical coke particles pack better than plate-like or needle-like ones, thus even particles with more porosity may result in a higher vibrated bulk density. Sverdlin et al. [6] reported that irregular shape and rough surface of coke particles up to 12 mm prevented the movement in a viscous medium. Edwards et al. [7] demonstrated that anode density increased with the addition of 20% of spherical shot coke to anode paste. Shot coke is an isotropic coke resembling small balls (<5 mm) and is produced through a different coking process than the traditional sponge ones [8]. Great improvement in baked apparent density was explained by higher apparent density and better packing properties of the spherical coke particles. The influence of particle shape and surface characteristics has been extensively studied for different particulate materials. In general, inter-particle friction increases with increasing the angularity, surface area and surface roughness of particles [9–11]. When the inter-particle friction increases, the particle bridging increases resulting in a higher resistance to rearrangement and a lower packing fraction of a powder [9,11–13].

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Dependence of anode properties on the paste formulation and the anode making parameters has been the principal subject of anode making literature. Little has been published on the influence of particle characteristics, more specifically of the particle shape. In anode manufacturing industry, different sources of coke are generally characterized by vibrated bulk density (VBD) in order to determine the pitch demand [14] and to evaluate the final density [1,15]. This work aimed at showing the influence of shape and density of coke particles on the compaction behavior and density of anode. Unlike the metallic powders where the particle density is known and does not change with particle size, density of porous coke particles is not known and varies with particle size, depending on the pore size and volume fraction. In this work, image analysis was used to determine the density and shape factors of coke particles for several size fractions, parameters that were not considered in the previous works available in the literature. They were then associated with the vibrated bulk density of particles, compaction behavior of anode paste and density of green anodes.

2. Materials and methods

Five calcined sponge cokes (A–E) and a calcined shot coke were used to make laboratory-scale anode samples. Table 1 shows the chemical composition and real density of the cokes. Table 2 presents the size fractions of coke and their proportions used in the anode paste formulation. A Nikon Ephiphot optical microscope equipped with an image analysis system (Clemex, vision) was used to measure five particle shape factors including aspect ratio, sphericity, roundness, compactness and convexity for every single fraction of the cokes. Definitions of the shape factors are given in Table 3. More spherical particles have lower aspect ratio and higher values for sphericity and roundness. Higher values of convexity and compactness show lower irregularity (angularity) of a particle. These factors describe various aspects of a particle, which may be associated with flow behavior and packing in a particle bed.

Apparent density of particles was determined by the analysis of optical microscopic images. Apparent density is defined as the ratio of the mass of a particle over its external envelope volume, which includes the internal pores. A known mass of particles was impregnated with resin under vacuum in a 2.5 cm diameter mold. The height of the impregnated samples was measured to calculate the sample volume. The samples were then sectioned vertically, as shown in Fig. 1, and the marked surfaces were polished. The area fraction of particles on the polished surfaces was determined by image analysis and averaged. The average area fraction was extrapolated to volume fraction and multiplied by the sample volume to obtain the apparent volume of coke particles only. Having the mass and volume of particles, apparent density was calculated.

For each fraction of coke the pycnometer density of particles with a batch weight of 40–55 g was measured by a He-pycnometer (Micromeritics, AccuPyc II 1340). Volume fractions of open and closed pores within the particles were calculated using Eqs. (2) and (3), respectively. Bulk density of single fractions of the coke particles was measured by a Scott volumeter (ASTM B329) using a 25 cm^3 cylindrical cup. This is called Scott density hereafter. Vibrated bulk density of each size fraction of the cokes was measured by a vibrating table (ASTM D4292). One hundred grams of particles was poured in a graduated cylinder while the cylinder was installed on a table and vibrated at 60 Hz with an amplitude of 0.2 mm. The volume of vibrated particles was read after 120 s of vibration. Void fraction between the vibrated particles was calculated according to Eq. (4).

$$\%\text{Total porosity} = \frac{\text{Real density} - \text{Apparent density}}{\text{Real density}} \times 100 \tag{1}$$

$$\%\text{Open porosity} = \frac{\text{Pycnometer density} - \text{Apparent density}}{\text{Pycnometer density}} \times 100 \tag{2}$$

$$\%\text{Closed porosity} = \%\text{Total porosity} - \%\text{Open porosity} \tag{3}$$

$$\%\text{Interparticle void} = \frac{\text{Apparent density} - \text{VBD}}{\text{Apparent density}} \times 100 \tag{4}$$

Angle of repose was also measured for each coke fraction according to ASTM C1444 standard method. The friction between particles is an indication of particle shape and their surface roughness.

A multivariate analysis was performed using Principal Component Analysis (PCA) method to determine the correlations between the shape factors of particles. PCA is a latent variable method with a wide range of applications and serves to reduce several variables into fewer numbers of latent variables or scores capturing most of the information contained in the original variables [16]. Latent variables are linear combination (regression) of the input variables as expressed in Eq. (5).

$$X = TP + E \tag{5}$$

where X is the matrix of input variables, P is the matrix of regression coefficients or loadings, T is the matrix of scores and E is the matrix of residuals. The loadings define the relative importance of each variable to the scores and are selected in such a way to describe the maximum variation in the variables.

Projection to Latent Structures (PLS) was implemented to predict the VBD from particle characteristics. PLS is another latent variable method, also known as partial least squares, that serves to predict the matrix of output parameters (Y) from the matrix of input variables (X) [16]. A score matrix for input variables (T) and a score matrix for output results (U) are defined with PCA. Then PLS tries to maximize the correlations between the T and U matrices using the appropriate loadings. Apparent density and particle shape factors were used as input variables in the PLS model to predict the VBD.

Two series of anode paste samples were made. For the first series, the size fractions of each sponge coke (Table 2) were mixed with pitch at a pitch to coke ratio of 16.2/100. The pitch had a Mettler softening point of 109 °C and a quinoline insoluble (QI) content of 15.5%. Pitch and coke fractions were individually weighed and added into the mixer to avoid variations in the composition of samples. Coke and pitch were preheated at 178 °C and mixed at the same temperature for 10 min [17]. The total mass of coke and pitch was 488 g for all samples. For the second series of samples, the −8 + 16 mesh size fraction of sponge cokes was replaced with the same size range of shot coke. Pitch to coke ratio and mixing parameters similar to the first series of pastes were applied.

Table 1
Chemical composition and real density of the cokes used for making anode samples.

Coke	Na [ppm]	Si [ppm]	%S	Ca [ppm]	V [ppm]	Fe [ppm]	Ni [ppm]	Real density (g/cm^3)
A	<50	100	1.1	130	120	70	90	2.075
B	<50	200	3.2	<10	260	60	150	2.063
C	<50	50	3.16	20	280	90	130	2.074
D	100	120	2.13	130	360	460	250	2.057
E	<50	210	1.73	240	90	340	230	2.073
Shot coke	140	75	3.85	37	1414	58	345	2.004