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New insight on the restructuring and breakage of particles during uniaxial confined compression tests on aggregates of petroleum coke

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ABSTRACT

With the aim of modelling the mechanical behaviour of carbon anode paste, compression tests within a rigid mould were performed on different recipes of carbon paste containing 32 to 44 wt.% binder. The compression was performed by applying a series of loading/unloading cycles. The maximum load was incrementally increased at each cycle. Test results showed that a softening behaviour occurs after the hardening of the anode paste during compaction cycles. This paper attempts to explain the mechanism of this peculiar behaviour. For comparison, two cokes with different shape factors were used to prepare the paste recipes. It was observed that round particles result in less resistance to local shear stress than do the angular ones. Binder, which consists of coal tar pitch and fine coke particles, acts as a lubricant enhancing the compaction of the paste. The breakage and slippage of particles during loading were monitored using an acoustic measurement system. A creep test, performed on a carbon paste and dry aggregates, corroborates that the time-dependency, or creep, of the paste is mainly governed by the solid skeleton.

1. Introduction

In the aluminium smelting process, carbon anodes are consumed in large quantities. Their production is an important part of the aluminium manufacturing process and a good quality anode helps to reduce both carbon and smelting energy consumptions. The anode paste is made of calcined petroleum coke, crushed recycled anode butts, coal tar pitch, and air voids. Coal tar pitch is solid at room temperature and becomes a viscous liquid at process temperatures, typically around 150 °C. The anode manufacturing process consists of three main stages: mixing, forming and baking. During the first stage, raw materials are mixed in a worm mixer with the aim of obtaining a uniform carbon paste mixture. In the second stage, the paste is poured into a rigid mould and either pressed or vibrocompacted. The formed anode block is then cooled down in a water pool allowing the green bloc to maintain its shape with sufficient mechanical properties required for further handling. The last stage consists in the baking of the green carbon block for about 20 days at a typical soaking temperature of 1200 °C [1].

The forming process of the green anode can be performed by pressing the carbon paste under a high uniaxial load or using a vibrocompaction process with significantly lower loading. The maximum vertical stress transmitted to the carbon paste during vibrocompaction has been estimated in this laboratory to be 3 MPa. The forming process lasts for about 65 s with a loading frequency around 25 Hz. The temperature of the paste is kept near 150 °C during the entire process.

During compaction, stress gradients through the paste results in considerable density gradients in the green anode blocs. The density gradient, in turn, affects the anode properties resulting in inhomogeneous electrical conductivity and chemical reactivity.

Numerical simulation of the forming process is a powerful tool to study the effect of different process and material variables on green density and to adjust certain process parameters in order to improve the anode quality. However, granular materials exhibit a non-linear anisotropic elasto-plastic behaviour making the development of the mechanical constitutive law, required to build a numerical model, rather complex [2]. There is a great resemblance between anode paste and asphalt mixture whose mechanical behaviour has widely been investigated. Rajagopal [3,4] proposed a macroscopic model based on a thermodynamic framework to predict the mechanical behaviour of asphalt mixtures. Although this model accurately predicts the behaviour of asphalt during the lay down (compaction) process, it does not take into account the mechanical transversal response such as the Poisson effect. Chaouki et al. [5] have used this model in the aim of predicting the carbon paste behaviour subjected to an axial loading. The results show that the model is also able to predict the evolution of the carbon paste density within a confined sample subjected to a high uniaxial stress.

Heckel [6,7] explained that the compaction process of a granular media, metallic powder for instance, occurs within three stages. In the first stage, the natural configuration of the paste is obtained after filling the mould. In the second stage, the individual rearrangement and

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rotation of particles are observed due to the increasing load applied on the granular media. Finally, after the load has reached a certain level, the compaction process is completed by an individual plastic deformation of the particles. Numerous authors investigated the compactibility of pharmaceutical powders based on Heckel's work. York [8] investigated the compactibility of lactose powders having a particle size smaller than 10 μm. His results show that particle slippage and rearrangement are the main causes of densification while particle crushing does not appear to have a significant contribution in densification. Similarly, Di Martino et al. [9] observed that brittle fractures have only a limited effect on densification of ibuprofen.

Unlike the compression of fine powders, where particles are more difficult to break due to their smallness, the crushing stage is much more important in the compaction of large granular materials. Fig. 1 presents the schematic representation of the compaction process of a granular material. Russell and Khalili [10] define the three stages differently to explain the compaction process of a granular media subjected to an increasing uniaxial load. Between points A and B, particles are rearranged by sliding and rotation to form a skeleton. At point B, an inflection in the curve separates stages I and II. For a petroleum coke, this inflection occurs around a specific volume 3.0 for a load neighbouring 0.65 MPa [11]. From point B to point C, particles are crushed and fragmented. The slope (λ_{cr}) is function of the aggregate size; the larger the aggregates, the steeper the slope [11,12]. Between points C and D, particle breakage is no longer the main compaction mechanism because the finer particle fragments become more difficult to break. At this stage, deformations are mainly conducted by an elastic behaviour. Many other researchers support the idea that the compaction process of a granular brittle material is composed of these three stages [11,12]. Brulin et al. [13] observed the same sequence of phenomena in their investigation related to the mechanical behaviour of ramming paste. The particularity of this material lies in its composition and texture which are similar to the anode carbon paste at 150 °C.

Many researchers have oriented their investigation towards creating a model that takes into account the three stages of granular material densification. Hagerty et al. [14] have studied the influence of particle shape using Ottawa sand, black beauty slag, and four different glass particles with a mean size ranging from 425 to 850 μm. They performed a high stress uniaxial compression test in a rigid die. Their investigations have mainly focused on the determination of a threshold corresponding to the beginning of the particle breakage. They observed that the transition between the first and second stages occurs at a higher stress level and a lower specific volume for the spherical Ottawa sand compared to the angular Black Beauty slag. It can be observed from this publication that the transition occurs at a specific volume of 1.6 and a load of 12.6 MPa for the sand. For the slag, it occurs at a specific volume of 1.75 and a load of 2.5 MPa. They concluded that angular particles start to break at a lower stress than do the spherical ones. They also showed that less crushing occurs for smaller glass beads. Similarly, within a study on rock crushing, Liu et al. [15] concluded that particle shape has a considerable influence on the breaking point threshold. Performing 2D simulations, they accurately captured the features of the breakage process.

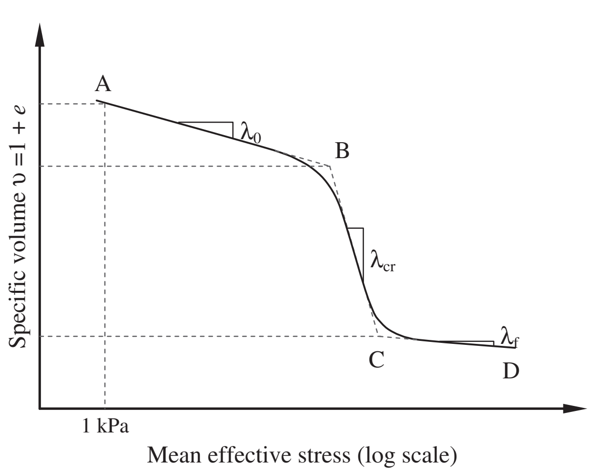

Fig. 1. Schematic representation of a granular material compaction [10].

To design a better constitutive model able to accurately predict the compaction behaviour of anode paste, the interaction between the aggregates as well as the main causes of inflection in deformation-stress curves should first be understood. To achieve these objectives, a simplified carbon paste recipe was chosen and a series of compaction experiments were performed. The particle distribution was limited to aggregates ranging between −8 and +14 US Mesh (1410 to 2380 μm). This particle selection has been made with the aim of reducing the impact of other phenomena that can bias the conclusions taken from the observations. The binder matrix, made of coal tar pitch and fine particles of coke, was kept as close as possible to the industrial recipe. Sponge coke and shot coke particles were used to investigate the influence of particle shape on compaction behaviour. Sponge coke particles have irregular shapes with asperities unlike shot coke particles, which are more spherical.

A first series of experiments focused on the influence of the binder content. Uniaxial compression tests were performed on pastes having 32 wt.% to 44 wt.% binder. Henceforth, the percentage and the fraction will be relative to the weight, unless otherwise specified. During these experiments, the pastes composed of sponge coke and shot coke have been subjected to the same trials and the influence of particle shape was revealed. In the second part of this investigation, similar experiments were performed on dry aggregates while the aggregate interactions were studied using an acoustic recording system. Finally, the time-dependant behaviour of compaction has been studied through a modified creep test using three loading levels. The coke was subjected to the creep test with and without binder in order to capture the origin of the time lag.

2. Materials and methods

2.1. Raw materials

Two types of coke aggregates were selected based on their shape. Sponge coke with irregular shape, which is mainly used in anode manufacturing, and shot coke, which has a rounder and more spherical shape. However, shot coke is usually not used in anode manufacture due to its high coefficient of thermal expansion (danger of anode cracking) and due to its high consumption rate in the smelting pot (more reactive). Roundness represents the angularity of the particle asperities. According to Wadell [16,17], roundness is the ratio of the average radius of curvature of the particle asperities to the radius of curvature of the largest circumscribed sphere. This definition gives lower values for rough particles and higher values for smooth ones independent of their sphericity. Sphericity roughly represents the particle shape. It is the ratio of the surface area of a sphere to the surface area of the particle with the same volume [16,17]. Fig. 2 shows a schematic representation of variations in particle shape.

Roundness and sphericity are important and useful concepts to explain particle interactions. A coke with low sphericity and roundness creates anchors between particles resulting in a considerable increase of the energy needed to cause a relative movement. This is due to the stronger friction force and energy needed to break the aggregates [18]. Fig. 3 shows the sponge coke and shot coke used for the experiments. One may observe that the sphericity and roundness of the shot coke are higher than those of sponge coke. The sphericity and roundness of the shot coke are respectively 0.863 and 0.745 compared to 0.730 and 0.622 for the sponge coke.

In order to obtain a paste, aggregates are bonded by means of a binder matrix. The binder was prepared by mixing fine coke powder with