VISCOPLASTIC MODELING OF THE GREEN ANODE FORMING PROCESS

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Abstract

To model the forming process of green anodes, a nonlinear viscoplastic constitutive law using the concept of natural configuration has been developed. For this purpose, a Helmholtz free energy was proposed in order to take into account the nonlinear compressible behaviour of the anode paste and a dissipation potential was introduced to characterize the irreversible deformation process. An experimental study based on compaction tests of the anode paste at 150 ^{o}C was carried out. To characterize the Poisson's effect, which leads to a non-negligible radial pressure, a thin steel instrumented mould was used. An inverse identification procedure using finite elements analysis showed that the model is able to reproduce experimental results in a good agreement.

Introduction

In the aluminium industry, the anode quality influences the efficiency of the Hall-Héroult electrolysis process. A poorly compacted carbon paste, with significant density gradients, could affect the electrical conductivity, the carbon consumption and may lead to initiation and propagation of cracks in the baked anode. Furthermore, the anode quality depends on several parameters like percentage of pitch, size of aggregates and temperature [1]. Thus, a high number of experimental tests are needed in order to optimize the forming process of the green anode. In a context where quality of the raw material could change having an impact on the final quality, the use of modelling and simulation becomes a good alternative to predict the density distribution and the evolution of the mechanical properties of the anode. However, these simulations must be based on a good knowledge of the material constitutive laws.

Research work dealing with the modelling of the anode paste compaction process, especially the development of constitutive laws, is very rare [2, 3]. However, considerable efforts have been done in order to study the mechanical behaviour of ramming paste and asphalt mixes. The composition of these materials and their compaction processes are similar, in some manner, to those of carbon paste.

Some works have investigated the modelling of asphalt mix compaction using the discrete element method [4, 5]. Such a technique is not suitable for large-scale engineering problems in comparison to macro-mechanical models. However, as in the given references, the technique may be used to enhance development of appropriate constitutive models.

Due to the irreversible deformation process characterizing the compaction of asphalt mixtures and ramming paste, several works using macroscopic material models were based on viscoplastic and elastoplastic behaviours. For ramming paste, a Cam-Clay model was used in [6]. For asphalt mixes, a viscoplastic model where Young modulus and viscosity are functions of strain rate was proposed by [7]; in [8], a viscoplastic constitutive model was developed where microstructural properties, such as microstructural orientation of aggregates, were related to the macroscopic model parameters; in [9], authors have proposed a new approach to develop a constitutive law, based on a the thermodynamic framework and the finite strain theory, where the concept of natural configuration is used to characterize the dissipation process [10].

This paper proposes a modified version of the material model proposed in [9] in order to simulate the compaction process of the anode paste using the finite element method. The constitutive law is described and the experimental results of the anode paste compaction test are presented. An inverse identification procedure, based on finite element analysis, shows that the model predicts the experimental trends.

Constitutive law

Let k_r be the reference configuration of the material and χ_{rt} the motion of the deformation. At a given time t, each point X of the reference configuration is mapped to a point $\mathbf{x} = \chi_{kr}(\mathbf{X}, \mathbf{t})$ belonging in the current configuration $k_{c(t)}$ (see Fig.1). One denotes $\mathbf{F}_{kr} = \nabla \chi_{kr}$ the deformation gradient tensor. Furthermore, let $\mathbf{C}_{kr} = \mathbf{F}_{kr}^{T}.\mathbf{F}_{kr}$ and $\mathbf{B}_{kr} = \mathbf{F}_{kr}.\mathbf{F}_{kr}^{T}$ the right and left Cauchy-Green stretch tensors, respectively. Let $\mathbf{L} = \mathbf{F}_{kr}.\mathbf{F}_{kr}^{-1}$ denote the velocity gradient and $\mathbf{D}$ its symmetric part.

From a mechanical point of view the forming process of the green anode is dissipative and irreversible. For this purpose, it would be appropriate to introduce the concept of the natural configuration, which corresponds at each time t to the stress-free state, denoted

by $k_{p(t)}$. Therefore, the deformation gradient tensor is decomposed multiplicatively as follows (see Fig.1):

$$F_{kr} = F_{k_{p(t)}} . G \qquad (1)$$

where $F_{k_{p(t)}}$ is the deformation gradient that accounts for the material's elastic response occurring between configurations $k_{p(t)}$ and $k_{c(t)}$. The tensor G characterizes the irreversible part of the deformation process.

To the natural configuration $k_{p(t)}$ one can associate: $C_{k_{p(t)}} = F_{k_{p(t)}}^T . F_{k_{p(t)}}$ the right Cauchy-Green tensor, $B_{k_{p(t)}} = F_{k_{p(t)}} . F_{k_{p(t)}}^T$ the left Cauchy-Green tensor, $L_{k_{p(t)}} = G.G^{-1}$ the velocity gradient and $D_{k_{p(t)}} = \frac{1}{2}\left[L_{k_{p(t)}} + L_{k_{p(t)}}^T \right]$ the rate of deformation tensor.
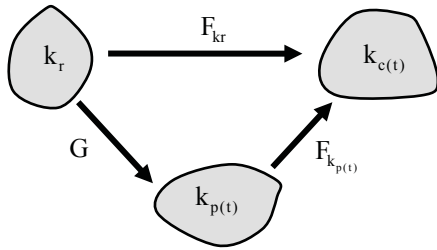

Figure 1. Natural configuration and deformation gradient tensor decomposition.

According to the decomposition of the gradient deformation tensor (eq.(1)), the material behaviour occurring between natural and current configuration is assumed to be elastic. Therefore, the state of stress will be characterized through elastic strain energy, defined with respect to the natural configuration.
The Clausius-Duhem inequality for an isothermal process is given by [11]:

$$\sigma : L - \rho \dot{\Psi} = \Phi \geq 0 \qquad (2)$$

where σ is the Cauchy stress tensor, Ψ the specific Helmholtz energy, Φ the mechanical energy dissipation related to the permanent strain and ρ is the density. It is assumed that the Helmholtz potential is a function of the following scalar invariants:

$$\Psi = \Psi\left(I_{B_{k_{p(t)}}}, III_{B_{k_{p(t)}}}, III_G \right) \qquad (3)$$

with

$$I_{B_{k_{p(t)}}} = Tr\left(B_{k_{p(t)}} \right), III_{B_{k_{p(t)}}} = \det\left(B_{k_{p(t)}} \right) \text{ and } III_G = \det(G) \qquad (4)$$

The scalar invariant III_G is related to the density evolution of the anode paste.

After some lengthy algebraic developments, the material derivative of Ψ is given by:

$$\dot{\Psi} = 2\frac{\partial\Psi}{\partial I_{B_{k_{p(t)}}}}\left(B_{k_{p(t)}} : D - C_{k_{p(t)}} : D_{k_{p(t)}} \right) +$$
$$2 III_{B_{k_{p(t)}}} \frac{\partial\Psi}{\partial III_{B_{k_{p(t)}}}}\left[I : \left(D - D_{k_{p(t)}} \right) \right] + \qquad (5)$$
$$III_G \frac{\partial\Psi}{\partial III_G} I : D_{k_{p(t)}}$$

Inserting this equation into the expression of the mechanical energy dissipation leads to:

$$0 \leq \Phi = \left[\sigma - 2\rho\frac{\partial\Psi}{\partial I_{B_{k_{p(t)}}}} B_{k_{p(t)}} - 2\rho III_{B_{k_{p(t)}}} \frac{\partial\Psi}{\partial III_{B_{k_{p(t)}}}} I \right] : D +$$
$$\left[2\rho\frac{\partial\Psi}{\partial I_{B_{k_{p(t)}}}} C_{k_{p(t)}} + 2\rho III_{B_{k_{p(t)}}} \frac{\partial\Psi}{\partial III_{B_{k_{p(t)}}}} I - \rho III_G \frac{\partial\Psi}{\partial III_G} I \right] : D_{k_{p(t)}} \qquad (6)$$

Assuming that the first term of Eq. (6) is related to the elastic behaviour of the material, the Cauchy stress tensor is thus given by:

$$\sigma = 2\rho\frac{\partial\Psi}{\partial I_{B_{k_{p(t)}}}} B_{k_{p(t)}} + 2\rho III_{B_{k_{p(t)}}} \frac{\partial\Psi}{\partial III_{B_{k_{p(t)}}}} I \qquad (7)$$

and the intrinsic energy dissipation becomes:

$$0 \leq \Phi = \rho\left(2\frac{\partial\Psi}{\partial I_{B_{k_{p(t)}}}} C_{k_{p(t)}} + 2 III_{B_{k_{p(t)}}} \frac{\partial\Psi}{\partial III_{B_{k_{p(t)}}}} I - III_G \frac{\partial\Psi}{\partial III_G} I \right) : D_{k_{p(t)}} \qquad (8)$$

According to the Eq.(7), the stress tensor is a function of kinematic variables related to the natural configuration, which is unknown. To complete the constitutive model, the natural configuration evolution must be characterized.
It is easily shown using Eq.(7) that the intrinsic energy dissipation can be rewritten as:

$$0 \leq \Phi = T : D_{k_{p(t)}} \qquad (9)$$

with

$$T = F_{k_{p(t)}}^T . \sigma . F_{k_{p(t)}}^{-T} - \rho III_G \frac{\partial\Psi}{\partial III_G} I \qquad (10)$$

Knowing that the dissipative energy must be positive, T can be defined as follows [12]:

$$\mathbf{T} = \eta\left(\mathrm{III}_G\right)\mathbf{C}_{k_{p(t)}}.\mathbf{D}_{k_{p(t)}} \tag{11}$$

Thus, the energy dissipation is defined as:

$$\Phi = \eta\left(\mathrm{III}_G\right)\mathbf{D}_{k_{p(t)}} : \left(\mathbf{C}_{k_{p(t)}} . \mathbf{D}_{k_{p(t)}}\right) \tag{12}$$

In the definition (11) the material parameter $\eta\left(\mathrm{III}_G\right)$ can be viewed as a viscosity function evolving with the anode paste compaction.

According to Eq.(10) and the definition (11) one obtains:

$$\mathbf{F}_{k_{p(t)}}^T.\boldsymbol{\sigma}.\mathbf{F}_{k_{p(t)}}^{-T} - \rho\,\mathrm{III}_G\,\frac{\partial\Psi}{\partial\mathrm{III}_G}\mathbf{I} = \eta\,\mathbf{C}_{k_{p(t)}}.\mathbf{D}_{k_{p(t)}} \tag{13}$$

Then, Cauchy stress and rate of deformation tensors are related through the formula:

$$\boldsymbol{\sigma} - \rho\,\mathrm{III}_G\,\frac{\partial\Psi}{\partial\mathrm{III}_G}\mathbf{I} = \eta\,\mathbf{F}_{k_{p(t)}}.\mathbf{D}_{k_{p(t)}}.\mathbf{F}_{k_{p(t)}}^T \tag{14}$$

Based on the material derivative of the tensor $\mathbf{B}_{k_{p(t)}}$, one can show that [9]:

$$\mathbf{F}_{k_{p(t)}}.\mathbf{D}_{k_{p(t)}}.\mathbf{F}_{k_{p(t)}}^T = -\frac{1}{2}\left(\overset{\bullet}{\mathbf{B}}_{k_{p(t)}} - \mathbf{L}.\mathbf{B}_{k_{p(t)}} - \mathbf{B}_{k_{p(t)}}.\mathbf{L}^T\right) = -\frac{1}{2}\overset{\triangledown}{\mathbf{B}}_{k_{p(t)}} \tag{15}$$

where $\overset{\triangledown}{\mathbf{B}}_{k_{p(t)}}$ is the upper convected Oldroyd derivative of $\mathbf{B}_{k_{p(t)}}$. Consequently, the evolution of the natural configuration is governed by the following differential equation:

$$\overset{\triangledown}{\mathbf{B}}_{k_{p(t)}} = -\frac{2}{\eta\left(\mathrm{III}_G\right)}\left(\boldsymbol{\sigma} - \rho\,\mathrm{III}_G\,\frac{\partial\Psi}{\partial\mathrm{III}_G}\mathbf{I}\right) \tag{16}$$

In the present work, the specific Helmholtz potential is considered as a neo-Hookean form for compressible material [11]:

$$\Psi\left(I_{B_{k_{p(t)}}}, \mathrm{III}_{B_{k_{p(t)}}}, \mathrm{III}_G\right) = \frac{\mu\left(\mathrm{III}_G\right)}{2\rho_{k_{p(t)}}}\left[I_{B_{k_{p(t)}}} - 3 - \ln\left(\mathrm{III}_{B_{k_{p(t)}}}\right)\right] + \frac{\lambda\left(\mathrm{III}_G\right)}{8\rho_{k_{p(t)}}}\ln\left(\mathrm{III}_{B_{k_{p(t)}}}\right)^2 \tag{17}$$

Therefore, the Cauchy stress tensor is rewritten as:

$$\boldsymbol{\sigma} = \frac{\mu\left(\mathrm{III}_G\right)}{\sqrt{\mathrm{III}_{B_{k_{p(t)}}}}}\mathbf{B}_{k_{p(t)}} + \frac{1}{\sqrt{\mathrm{III}_{B_{k_{p(t)}}}}}\left(\frac{\lambda\left(\mathrm{III}_G\right)}{2}\ln\left(\mathrm{III}_{B_{k_{p(t)}}}\right) - \mu\left(\mathrm{III}_G\right)\right)\mathbf{I} \tag{18}$$

Functions $\mu(.)$ and $\eta(.)$ can be viewed respectively as shear modulus and viscosity function. They evolve as well as the material is compacted.

To simulate the compaction process of green anode, the following expressions of shear and viscosity functions will be used [9]:

$$\mu\left(\mathrm{III}_G\right) = \hat{\mu}\left[1 + \lambda_1\left(\mathrm{III}_G\right)^{2n_1}\right]^{q_1} \tag{19}$$

$$\eta\left(\mathrm{III}_G\right) = \hat{\eta}\left[1 + \lambda_2\left(\mathrm{III}_G\right)^{2n_2}\right]^{q_2} \tag{20}$$

where $\hat{\mu},\hat{\eta},\left(\lambda_i\right)_{i=1,2},\left(n_i\right)_{i=1,2}$ and $\left(q_i\right)_{i=1,2}$ are material parameters. The function $\lambda(.)$ is related to the Poisson's effect and it is defined as follows:

$$\lambda\left(\mathrm{III}_G\right) = \alpha\,\exp\left(\frac{1 - \mathrm{III}_G}{\beta}\right) \tag{21}$$

where α and β are material parameters.

In [3] and [9] the parameter function λ was set equal to zero. In such a case the material model cannot capture the Poisson's effect.

Experimental study and inverse identification procedure

In order to identify parameters of the proposed constitutive law and to verify its predictive capability, compaction tests at 150ºC were performed with anode paste. For this purpose, a thin wall mould, having a height of 140 mm, a diameter of 254 mm and a wall thickness of 0.356 mm, was used. The mould was instrumented in order to measure both axial and radial stresses applied on the sample. The axial stress is easily measured trough the load cell. For the radial stress, circumferential and axial gauges were set around the die to evaluate average circumferential and axial strains of the mould's wall (see Fig.2). Thus, using the theory of linear elasticity for a thin shell submitted to internal pressure, one can obtain the radial stress applied on the anode paste. Such an approach provides also an estimation of the radial displacement of the anode paste or the mould's wall. For more details on experimental setup and the estimation of this radial stress, acting on the anode paste, the reader is referred to [13]. The anode paste mixture consists of a commercial anode-grade coke and pitch (16%).

Figure 2. Experimental setup.

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To identify material parameters a user's material subroutine VUMAT for an explicit dynamic analysis has been developed and implemented in ABAQUS software. The purpose of the simulation is to demonstrate the ability of the proposed constitutive law to predict experimental trends.

Figure 3 shows the CAD model used. Due to confidentiality, material parameters used for finite element simulation will not be disclosed.

Figures 4-8 show a benchmark between experimental and finite element results. The proposed constitutive law predicts quite successfully axial pressure acting on the anode paste (Fig. 4), circumferential strain (Fig.5) and radial displacement (Fig.6) of the mould's wall and the apparent density (Fig. 8). For the radial pressure, the numerical prediction is slightly overestimated at the end of the test compared to the experimental result (Fig. 7).

Notice that the axial pressure reached approximately 4 MPa at the end of the test and the paste was approximately compacted up to 35 % of its initial volume. Furthermore, the estimated radial pressure is approximately half of the axial pressure. Consequently, it is clear that the Poisson's effect cannot be neglected during the compaction process.
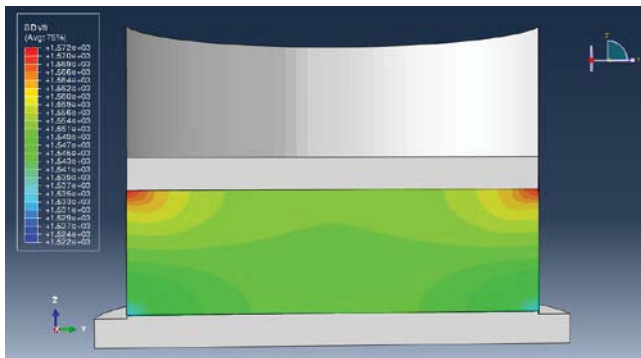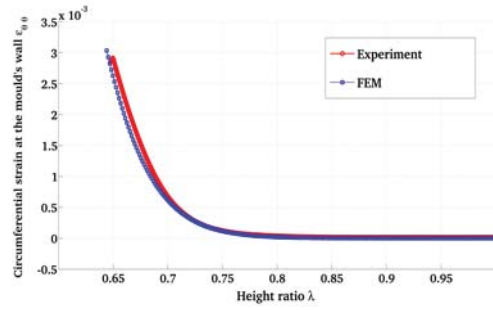

Figure 5. Circumferential strain at the mould's wall: FEM vs experiment.

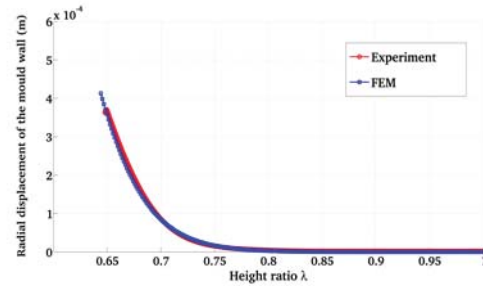Figure 6. Radial displacement at the mould's wall: FEM vs experiment.

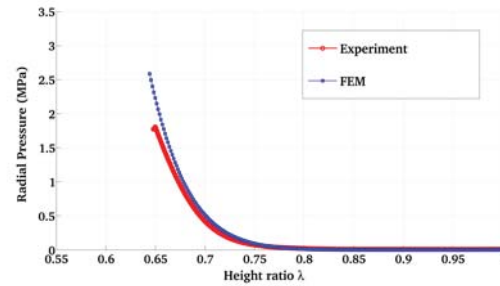Figure 3. CAD model.

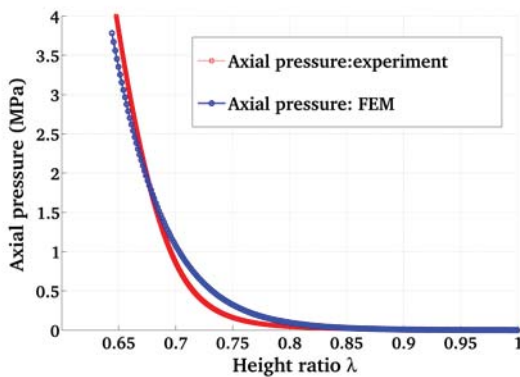Figure 7. Radial pressure: FEM vs experiment.

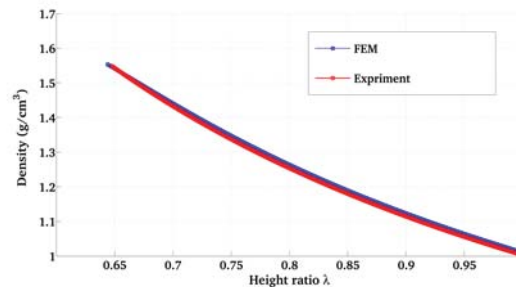Figure 4. Axial pressure: FEM vs experiment.

Figure 8. The apparent density: FEM vs experiment.

CONCLUSION

In this work a non-linear macroscopic viscoplastic constitutive law is used in order to simulate the anode paste compaction process. For this purpose, a thermodynamic formulation based on the natural configuration concept is used. The stress tensor is expressed as a function of a strain tensor defined with respect to the natural configuration. A dissipation potential is proposed to characterize the evolution of this natural configuration.

Simple compaction tests were carried out with a thin mould wall in order to characterize the mechanical behaviour of the anode paste during the compaction process. The mould was instrumented to provide both axial and radial stresses. An inverse identification procedure based on finite element analysis shows that the proposed model reproduces experimental trends in a good agreement.

However, some aspects, closely related to the compaction process, are still unclear, especially the contact between the anode paste and the mould and thermal effects. Moreover, further work is needed for a better understanding of the physical meaning of the material parameters used in the constitutive law.

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