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Michaël Peigney. A Melan theorem in diffusion-induced plasticity: Applications to lithium-ion batteries. 4th Polish Congress of Mechanics and the 23rd International Conference on Computer Methods in Mechanics, 2019, Cracovie, Poland. pp.020041, 10.1063/5.0007995 . hal-02985289

**HAL Id: hal-02985289**

**<https://enpc.hal.science/hal-02985289>**

Submitted on 2 Nov 2020

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Cite as: AIP Conference Proceedings **2239**, 020041 (2020); <https://doi.org/10.1063/5.0007995>  
Published Online: 22 May 2020

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# A Melan theorem in diffusion-induced plasticity: applications to lithium-ion batteries

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**Abstract.** We present a Melan-type theorem applying to cyclically loaded media in which plastic flow and diffusion of guest atoms are coupled, such as electrode materials in lithium-ion batteries. The presented theorem ensures that elastic shakedown occurs ( i.e. that the plastic strain stabilizes to a time-independent limit), independently of the initial state. Building on that theorem, a general two-step procedure is presented for designing any given structure against elastic shakedown. As an illustration, that procedure is applied to the model problem of a spherical electrode particle under time-harmonic lithiation.

## INTRODUCTION

Electrode materials in lithium-ion batteries are an example of medium in which stress and plastic flow are generated by the diffusion of guest atoms. In such media, both elastic and plastic deformations are strongly coupled with diffusion. For a free-standing electrode particle, the yield limit can, for instance, be reached by applying a purely chemical loading at a sufficiently high charging rate [1]. This raises the issue of studying how plasticity and diffusion evolve over consecutive loading cycles. For a certain range of cyclic loadings, numerical simulations reported on a spherical geometry show that elastic shakedown occurs, i.e. that the plastic strain stops evolving after a few cycles, so that the particle evolves elastically in subsequent cycles. Compared to other possible cyclic regimes such as alternate plasticity, elastic shakedown is arguably the most beneficial both with respect to fatigue [2] and with respect to the electrochemical efficiency [1].

In standard plasticity (without diffusion), Melan theorem gives a sufficient condition on the loading for ensuring that elastic shakedown occurs [3, 4]. A salient feature of that theorem is that it is path-independent, i.e. independent of any residual stress that may exist in the initial state. Melan theorem has been extended to several more complex behaviors than perfect plasticity, see e.g. [5, 6, 7, 8, 9, 10, 11] for relatively recent examples.

This paper reports on an extension of Melan's theorem to continuous media in which plasticity and diffusion are coupled. The problem is formulated in the geometrically linear setting and appeals to the consistent thermodynamic framework originally introduced by [12]. A Melan theorem for the considered media is presented, leading to a general two-step procedure for designing any given structure against elastic shakedown. An application to the model problem of a spherical electrode particle under cyclic lithiation-delithiation is presented. Explicit expressions are obtained. For the reader's convenience, a list of the main symbols is reported in Table I.

**TABLE I.** List of symbols.

Symbol	Description	Unit
$\varepsilon$	total strain	-
$\varepsilon^p$	plastic strain	-
$\sigma$	stress	Pa
$c$	concentration of guest atoms	$\text{m}^{-3}$
$j$	flux of guest atoms	$\text{m}^{-2}.\text{s}^{-1}$
$\mu$	chemical potential	J
$\mathbb{L}$	elasticity tensor	Pa
$A$	coupling parameter	J
$\mathcal{C}$	elasticity domain	-
$\psi$	diffusion potential	$\text{Pa}.\text{m}^{-3}.\text{s}^{-1}$

# DIFFUSION-INDUCED PLASTICITY IN A CONTINUUM

## Constitutive equations

An elastic-plastic constitutive material is considered. The local state of the material is described by the total linearized strain  $\varepsilon$ , the (deviatoric) plastic strain  $\varepsilon^p$  and the concentration  $c$  of guest atoms. The free energy  $w$  of the material is taken as a quadratic function of the form

$$w(\varepsilon, \varepsilon^p, c) = \frac{1}{2}(\varepsilon - \varepsilon^p) : \mathbb{L} : (\varepsilon - \varepsilon^p) + \frac{1}{2}kc^2 + cA : (\varepsilon - \varepsilon^p) + c\mu_0 \quad (1)$$

where  $\mathbb{L}$  is the symmetric positive definite fourth-order elasticity tensor and  $A$  is a symmetric second-order tensor accounting for the coupling between the elastic strain  $\varepsilon - \varepsilon^p$  and the concentration  $c$ . The material parameter  $k$  is assumed to satisfy the relation  $k > A : \mathbb{L}^{-1} : A$  which ensures that the function  $w$  in (1) is convex.

From (1) we obtain the expressions of the stress  $\sigma$  and the chemical potential  $\mu$  as

$$\begin{aligned} \sigma &= \frac{\partial w}{\partial \varepsilon} = \mathbb{L} : (\varepsilon - \varepsilon^p) + cA, \\ \mu &= \frac{\partial w}{\partial c} = \mu_0 + kc + A : (\varepsilon - \varepsilon^p). \end{aligned} \quad (2)$$

Observe that the concentration  $c$  has a linear influence on the stress, in a way similar to thermal stress. Similarly, the chemical potential  $\mu$  depends linearly on the elastic strain  $\varepsilon - \varepsilon^p$ .

The constitutive equations (2) are complemented by a law of diffusion and a plasticity flow rule. The flux  $j$  of guest atoms is assumed to obey the relation

$$j = -\partial \psi(\nabla \mu). \quad (3)$$

where  $\psi$  is differentiable and strictly convex. A normality flow rule is assumed for the plastic behavior, i.e.

$$\dot{\varepsilon}^p = \lambda \frac{\partial f}{\partial \sigma}(\sigma) \text{ with } \lambda \geq 0, f(\sigma) \leq 0, \lambda f(\sigma) = 0. \quad (4)$$

where  $f$  is a differentiable, strictly convex function of the deviatoric stress. We denote by  $\mathcal{C}$  the elasticity domain of the material, i.e.

$$\mathcal{C} = \{\sigma : f(\sigma) \leq 0\}.$$

## Conservation equations

Consider a deformable continuum occupying a domain  $\Omega$ , in which guest atoms diffuse. A flux  $J$  of guest atoms is prescribed on a part  $\Gamma_J$  of the boundary. On  $\Gamma_\mu = \partial\Omega - \Gamma_J$ , the chemical potential  $\mu$  is prescribed to take a given value  $M$ . Tractions  $T$  are prescribed on a part  $\Gamma_T$  of the boundary and displacements  $U$  are prescribed on  $\Gamma_u = \partial\Omega - \Gamma_T$ .

The mass conservation leads to the diffusion equation

$$\dot{c} + \text{div } j = 0 \text{ in } \Omega. \quad (5)$$

Equation (5) is complemented by the boundary conditions

$$\mu = M \text{ on } \Gamma_\mu, \quad j \cdot n = J \text{ on } \Gamma_J \quad (6)$$

where  $n$  is the outward-pointing normal vector.

Assuming deformation to be much faster than diffusion, the stress field  $\sigma$  satisfies the equilibrium equation

$$\text{div } \sigma + f = 0 \text{ in } \Omega \quad (7)$$

where  $f$  is the body force. Eq. (7) is complemented by the boundary conditions

$$\sigma \cdot n = T \text{ on } \Gamma_T, \quad u = U \text{ on } \Gamma_u \quad (8)$$

where  $u$  is the displacement field. The terms  $M, J, f, U$  and  $T$  are functions of the location  $x$  and time  $t$ . Similarly, the fields  $c, \mu, u$  and  $\sigma$  are all functions of  $x$  and  $t$ . Unless in the case of possible ambiguities, the dependence in  $(x, t)$  will be omitted in the notations.

The set of equations (2), (3), (4), (5), (6), (7) and (8) is denoted by  $(\mathcal{E})$  in the following. It governs the evolution of the continuum starting from a given initial values of the plastic strain and concentration fields.

## SHAKEDOWN DESIGN

From now on, attention is restricted to cyclic chemomechanical loadings, i.e. the functions  $M, J, f, U$  and  $T$  are all periodic in time (with the same period  $T$ ). Functions that are periodic in time with a period  $T$  are referred to as  $T$ -periodic in the following.

Let  $(u^E, \sigma^E, c^E, \mu^E)$  denote a cyclic steady state elasto-diffusive solution, i.e.  $(u^E, \sigma^E, c^E, \mu^E)$  are  $T$ -periodic functions satisfying

$$\begin{aligned} \sigma^E &= \mathbb{L} : \varepsilon^E + c^E A, \\ \mu^E &= \mu_0 + kc^E + A : \varepsilon^E, \\ \varepsilon^E &= \frac{1}{2}(\nabla u^E + \nabla^t u^E), \\ j^E &= -\partial \psi(\nabla \mu^E), \\ c^E + \operatorname{div} j^E &= 0 \text{ in } \Omega, \\ \operatorname{div} \sigma^E + f &= 0 \text{ in } \Omega, \end{aligned} \tag{9}$$

as well as the boundary conditions (6) and (8). Problem (9) amounts to discard plasticity in the constitutive laws. The existence of a  $T$ -periodic solution to (9) has been proved in [13]. Regarding the original problem  $(\mathcal{E})$  in which plasticity and diffusion are coupled, the following result can be proved [13]:

*If there exists a time-independent self-equilibrated [14] stress field  $\rho$  such that  $\sigma^E(x, t) + \rho(x)$  is in the interior of  $\mathcal{C}$  for all  $x \in \Omega$  and all  $t \in [0, T]$ , then shakedown occurs whatever the initial state.*

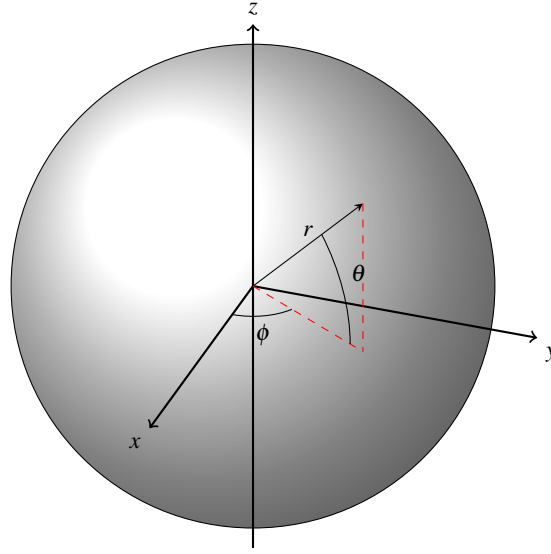
This theoretical result suggests a general two-step procedure for designing structures against elastic shakedown, in the framework of coupled plasticity-diffusion:

1. Calculate a  $T$ -periodic solution  $(u^E, \sigma^E, c^E, \mu^E)$  to the purely elasto-diffusive problem (9).
2. Check if there exists a time-independent self-equilibrated stress field  $\rho$  such that  $\sigma^E(x, t) + \rho(x)$  is in the interior of  $\mathcal{C}$  for all  $x \in \Omega$  and all  $t \in [0, T]$

Even though the purely elasto-diffusive problem (9) is simpler to handle than the original problem  $(\mathcal{E})$ , performing Step 1. above is not entirely trivial in the general case of a nonlinear diffusion law. Optimal control strategies [15, 16, 17] could possibly be useful in that case. The situation simplifies significantly when the diffusion law is linear, i.e. Fick's law for diffusion is considered. This correspond to the case where the diffusion potential  $\psi$  is quadratic. In such case, the purely elasto-diffusive problem (9) is linear. A Fourier decomposition can be used for constructing a  $T$ -periodic solution to (9).

Regarding Step 2., the crucial observation is that it is identical to the application of Melan theorem in standard plasticity (without diffusion). All the existing algorithms proposed for standard plasticity [18] can thus be used directly. In practice, Step 2. is performed by considering a finite dimensional space of self-equilibrated fields. This generally results in lower bounds on the loading parameters for which shakedown occurs.

*Remark:* The aforementioned theorem can extended to viscoplasticity, i.e. to the case where the flow rule reads as  $\dot{\varepsilon}^p = \partial g(s)$  where  $g$  is a differentiable, convex function of the deviatoric stress  $s$ . In such case, the condition that " $\sigma^E(x, t) + \rho(x)$  is in the interior of  $\mathcal{C}$ " in the statement of the theorem can be relaxed as " $\sigma^E(x, t) + \rho(x) \in \mathcal{C}$ ". Step 2 is modified accordingly.



**FIGURE 1.** Spherical particle.

## APPLICATION: SPHERICAL ELECTRODE PARTICLE

We consider the model problem of a free-standing spherical electrode particle with radius  $R$  (Fig. 1). Cyclic lithiation-delithiation is achieved by applying a  $T$ -periodic flux

$$J(t) = -\frac{R\omega}{6}H \sin \omega t \quad (10)$$

on the boundary. In (10),  $\omega$  and  $H > 0$  are given. The loading parameter  $H$  in (10) is directly related to the maximum number of inserted lithium ions. More precisely, the maximum number of inserted lithium ions is given by  $-4\pi R^2 \int_0^{T/2} J(t) dt = \frac{4}{3}\pi R^3 H$  which corresponds to an average concentration equal to  $H$ .

A hypothetical elastic-perfectly plastic material is considered. The free energy  $w$  is taken as

$$w(\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^p, c) = \frac{1}{2}K(tr \boldsymbol{\varepsilon})^2 + G(\boldsymbol{\varepsilon}^d - \boldsymbol{\varepsilon}^p) : (\boldsymbol{\varepsilon}^d - \boldsymbol{\varepsilon}^p) + \frac{1}{2}kc^2 + a tr \boldsymbol{\varepsilon} + c\mu_0 \quad (11)$$

where  $\boldsymbol{\varepsilon}^d$  is the deviatoric strain and  $c$  is the concentration of lithium. In (11),  $K$ ,  $G$ ,  $a$  and  $\mu_0$  are material parameters. The constitutive relations (2) are

$$\begin{aligned} \sigma_m &= K tr \boldsymbol{\varepsilon} + ac, \quad s = 2G(\boldsymbol{\varepsilon}^d - \boldsymbol{\varepsilon}^p), \\ \mu &= \mu_0 + kc + a tr \boldsymbol{\varepsilon}, \end{aligned} \quad (12)$$

where  $\sigma_m = (tr \boldsymbol{\sigma})/3$  is the hydrostatic stress and  $s$  is the deviatoric stress. The scalar material parameter  $a$  in (11) accounts for the chemo-mechanical coupling. In particular, for stress-free states in the elastic regime, Eq. (12) gives

$$tr \boldsymbol{\varepsilon} = -\frac{a}{K}c$$

hence the ratio  $-a/K$  can be interpreted as the volumetric expansion coefficient associated with the insertion of lithium.

Fick's law is adopted for diffusion, i.e.

$$\mathbf{j} = \frac{D}{k} \nabla \mu$$

where  $D$  is the diffusion coefficient.

The elasticity domain  $\mathcal{C}$  is of the von Mises type, i.e. defined by

$$\frac{1}{2}s : s \leq \sigma_Y^2 \quad (13)$$

where  $\sigma_Y$  is the yield strength.

In the following, the two-step procedure presented previously is applied for finding the range of loading parameters  $(\omega, H)$  for which shakedown occurs.

### Purely elasto-diffusive response

The first step consists in finding a  $T$ -periodic solution  $(\sigma^E, \mu^E, u^E, c^E)$  to the elasto-diffusive problem (9). For the harmonic loading (10), such a  $T$ -periodic solution  $(\sigma^E, \mu^E, u^E, c^E)$  can be sought in the form

$$\begin{aligned} \sigma^E(x, t) &= \Re(\hat{\sigma}^E(x)e^{i\omega t}), \quad \mu^E(x, t) = \mu_0 + \Re(\hat{\mu}^E(x)e^{i\omega t}), \quad j^E(x, t) = \Re(\hat{j}^E(x)e^{i\omega t}), \\ u^E(x, t) &= \Re(\hat{u}^E(x)e^{i\omega t}), \quad c^E(x, t) = \Re(\hat{c}^E(x)e^{i\omega t}), \end{aligned} \quad (14)$$

Here and in the following, the superscript  $\hat{\cdot}$  is used to denote complex-valued quantities. The real and imaginary parts of complex-valued quantities are denoted by  $\Re$  and  $\Im$ , respectively.

In view of the spherical symmetry of the problem, we look for a solution in the form

$$\hat{u}(x) = \hat{u}^E(r)e_r, \hat{c}(x) = \hat{c}^E(r)$$

where  $(e_r, e_\theta, e_\phi)$  is the local basis for the spherical coordinates  $(r, \theta, \phi)$ . The constitutive relations (12) show that

$$\begin{aligned} \hat{j}^E &= -(D/k) \frac{d\hat{\mu}^E}{dr} e_r, \\ \hat{\sigma}^E &= \hat{\sigma}_r^E e_r \otimes e_r + \hat{\sigma}_\theta^E (e_\theta \otimes e_\theta + e_\phi \otimes e_\phi), \end{aligned} \quad (15)$$

where

$$\begin{aligned} \hat{\mu}^E &= k\hat{c}^E + a \left( \frac{d\hat{u}^E}{dr} + 2\frac{\hat{u}^E}{r} \right), \\ \hat{\sigma}_r^E &= \frac{4G}{3} \left( \frac{d\hat{u}^E}{dr} - \frac{\hat{u}^E}{r} \right) + K \left( \frac{d\hat{u}^E}{dr} + 2\frac{\hat{u}^E}{r} \right) + a\hat{c}^E, \\ \hat{\sigma}_\theta^E &= \frac{2G}{3} \left( \frac{\hat{u}^E}{r} - \frac{d\hat{u}^E}{dr} \right) + K \left( \frac{d\hat{u}^E}{dr} + 2\frac{\hat{u}^E}{r} \right) + a\hat{c}^E. \end{aligned} \quad (16)$$

The functions  $\hat{c}^E$  and  $\hat{u}^E$  are obtained by solving the partial differential equations

$$\operatorname{div} \hat{\sigma}^E = 0, \operatorname{div} \hat{j}^E + i\omega \hat{c}^E = 0 \quad (17)$$

with the boundary conditions  $\hat{\sigma}_r^E = 0$  and  $e_r \cdot \hat{j}^E = iR\omega H/6$  at  $r = R$ . The solution to this boundary value problem can be expressed in terms of the spherical Bessel functions  $j_0$  and  $j_1$  defined on  $\mathbb{C}^*$  by

$$j_0(z) = \frac{\sin z}{z}, \quad j_1(z) = \frac{\sin z}{z^2} - \frac{\cos z}{z}.$$

Setting

$$\hat{\lambda} = e^{i\frac{3\pi}{4}} \sqrt{\frac{\omega}{D(1 - a\tilde{a}/k)}}, \quad \tilde{a} = \frac{3a}{3K + 4G}, \quad (18)$$

the solution is indeed found to be given by

$$\hat{c}^E = -\frac{H}{6} \frac{\hat{R} j_0(\hat{r})}{j_1(\hat{R})}, \quad \frac{\hat{u}^E}{R} = \tilde{a} \frac{H}{6} \left( \frac{4G}{3K} \frac{\hat{r}}{\hat{R}} + \frac{j_1(\hat{r})}{j_1(\hat{R})} \right) \quad (19)$$

where  $\hat{r} = \hat{\lambda}r$  and  $\hat{R} = \hat{\lambda}R$ . Replacing in (16) gives the expression of the stress  $\hat{\sigma}$  as

$$\begin{aligned}\hat{\sigma}_r^E &= \frac{2G\tilde{a}H}{3} \left( 1 - \frac{\hat{R}}{\hat{r}} \frac{j_1(\hat{r})}{j_1(\hat{R})} \right), \\ \hat{\sigma}_\theta^E &= \frac{2G\tilde{a}H}{3} \left( 1 + \frac{\hat{R}}{\hat{r}} \frac{j_1(\hat{r})}{2j_1(\hat{R})} - \frac{\hat{R}j_0(\hat{r})}{2j_1(\hat{R})} \right),\end{aligned}\tag{20}$$

as well as the expression of the chemical potential  $\hat{\mu}$  as

$$\hat{\mu}^E = \frac{H}{6} \left( (a\tilde{a} - k) \frac{\hat{R}j_0(\hat{r})}{j_1(\hat{R})} + \frac{4G}{K} a\tilde{a} \right).\tag{21}$$

Let  $s^E$  be the deviatoric part of  $\sigma^E$ . The obtained expressions for  $\sigma^E$  show that

$$s^E(x, t) = s_\theta^E(r, t) (-2e_r \otimes e_r + e_\theta \otimes e_\theta + e_\phi \otimes e_\phi)\tag{22}$$

where  $s_\theta^E(x, t) = \Re(\hat{s}_\theta^E(r) e^{i\omega t})$  and

$$\hat{s}_\theta^E(r) = \frac{GH\tilde{a}}{3} \frac{\hat{R}}{j_1(\hat{R})} \left( \frac{j_1(\hat{r})}{\hat{r}} - \frac{j_0(\hat{r})}{3} \right).\tag{23}$$

As an illustration,  $|\hat{s}_\theta^E(r)|$  is shown in Fig. 2 as a function of  $r/R$  for several values of the dimensionless charging rate  $\tilde{\omega}$  defined as

$$\tilde{\omega} = \Omega T_0\tag{24}$$

where

$$T_0 = \frac{R^2}{D \left( 1 - \frac{a\tilde{a}}{k} \right)}.$$

Since  $s_\theta^E(x, t) = \Re(\hat{s}_\theta^E(r) e^{i\omega t})$ , the function  $|\hat{s}_\theta^E(r)|$  corresponds to the time amplitude of the variation of  $s_\theta^E(x, t)$  at any given location  $x$ . The relation (22) also shows that  $\frac{1}{2}s^E : s^E = 3(s_\theta^E : s_\theta^E)^2$ . Hence the function  $|\hat{s}_\theta^E(r)|$  in Fig. 2 is directly connected to the maximum value (with respect to time) of the von Mises stress.

## Application of the shakedown condition

Having now found a  $T$ -periodic solution  $(\sigma^E, \mu^E, u^E, c^E)$  to (9), the second step consists in looking for a time-independent stress-field  $\rho$  such that

$$\frac{1}{2}(s^E(x, t) + \rho^d(x)) : (s^E(x, t) + \rho^d(x)) < \sigma_Y^2\tag{25}$$

for all  $x$  and for all  $t$ . In (25),  $\rho^d$  is the deviatoric part of  $\rho$ . For our purpose, it is sufficient to take  $\rho = 0$ , in which case the requirement (25) reduces to

$$3(s_\theta^E(x, t))^2 < \sigma_Y^2\tag{26}$$

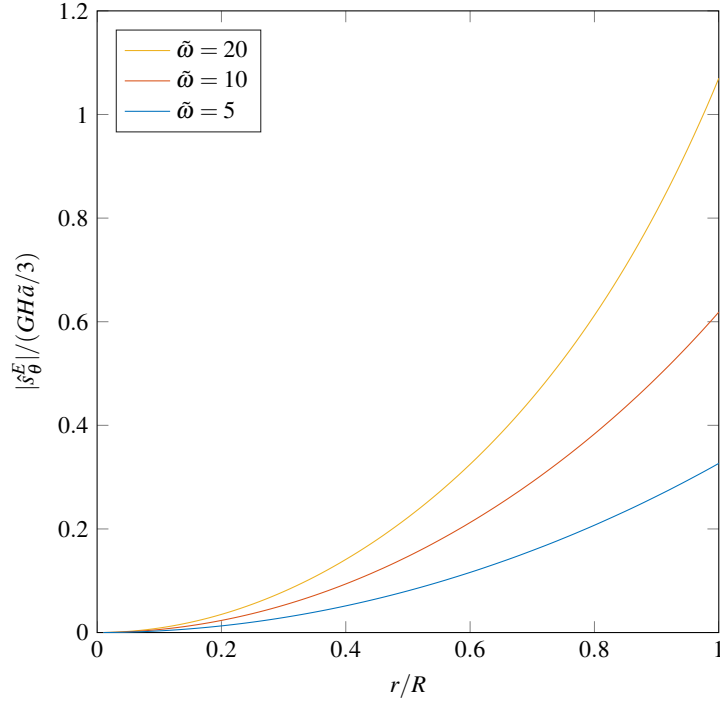
for all  $x$  and for all  $t$ . Since  $s_\theta^E(x, t) = \Re(\hat{s}_\theta^E(r) e^{i\omega t})$ , we have  $\sup_t (s_\theta^E(x, t))^2 = |\hat{s}_\theta^E(r)|^2$  so that condition (26) reduces to

$$3|\hat{s}_\theta^E(r)|^2 < \sigma_Y^2\tag{27}$$

for all  $r$ . An important observation is that  $|\hat{s}_\theta^E(r)|$  is maximum at  $r = R$ , see Fig. 2. Condition (27) thus reduces to  $|\hat{s}_\theta^E(R)| < \sqrt{\frac{1}{3}}\sigma_Y$ . From (23) we have

$$|\hat{s}_\theta^E(R)| = \frac{GH|\tilde{a}|}{9} \sqrt{(\Re z - 3)^2 + (\Im z)^2}$$





**FIGURE 2.** Cyclic steady state for a spherical electrode particle in the elastic shakedown regime: Time amplitude of the hoop deviatoric stress as a function on the location  $r$ , for several values of the charging rate  $\tilde{\omega}$ .

where

$$z = \frac{\hat{R}j_0(\hat{R})}{j_1(\hat{R})}.$$

Condition (25) finally reads as  $H < H_{SD}(\omega)$  where

$$H_{SD}(\omega) = 3 \frac{\sqrt{3}\sigma_Y}{G|\tilde{a}|} \left( (\Re z - 3)^2 + (\Im z)^2 \right)^{-\frac{1}{2}}$$

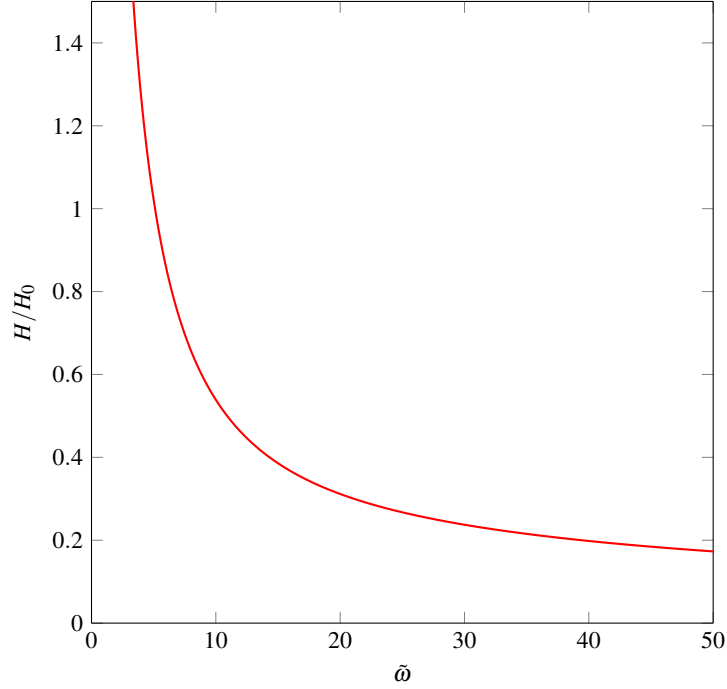
Setting

$$H_0 = 3 \frac{\sqrt{3}\sigma_Y}{G|\tilde{a}|} \quad (28)$$

the shakedown limit  $H_{SD}/H_0$  is shown in Fig. 3 in terms of the dimensionless charging rate  $\tilde{\omega}$ .

Whatever the initial state, shakedown occurs for any loading parameters  $(\tilde{\omega}, H)$  below the curve shown in Fig. 3. The charging rate  $\tilde{\omega}$  has a detrimental effect on the shakedown behavior: the higher the charging rate, the lower the number of lithium ions that can be injected for the particle to remain in the shakedown regime. This behavior results from a stress concentration phenomenon: for high charging rate, the injected ions do not have time to diffuse in the whole particle and remain trapped in a spherical shell near the boundary. Such a concentration of those injected ions generates large swelling near the boundary, which in turns generates high stress as illustrated in Fig. 2. In contrast, in the limit  $\omega \rightarrow 0$ , the lithium ions diffuse uniformly in the whole particle, so that the particle experiences a uniform stress-free swelling. Accordingly, the shakedown limit  $H_{SD}(\omega)$  tends to infinity as  $\tilde{\omega}$  tends towards 0.

For Li-ion batteries, a quantity of interest is the charge–voltage curve, showing the total charge  $C = \int_{\Omega} c d\Omega$  (or the average concentration  $\bar{C}/|\Omega|$ ) versus the voltage  $V = -(\mu - \mu_0)e$  on the surface (here  $e = 1.602 \times 10^{-19}$  C is the elementary charge). If the loading parameters  $(\tilde{\omega}, H)$  respect the shakedown condition  $H \leq H_{SD}(\omega)$ , the particle reaches a cyclic steady state in which in particular the chemical potential  $\mu$  and the local concentration  $c$  differ from the



**FIGURE 3.** Shakedown limit for a spherical electrode particle under cyclic lithiation.

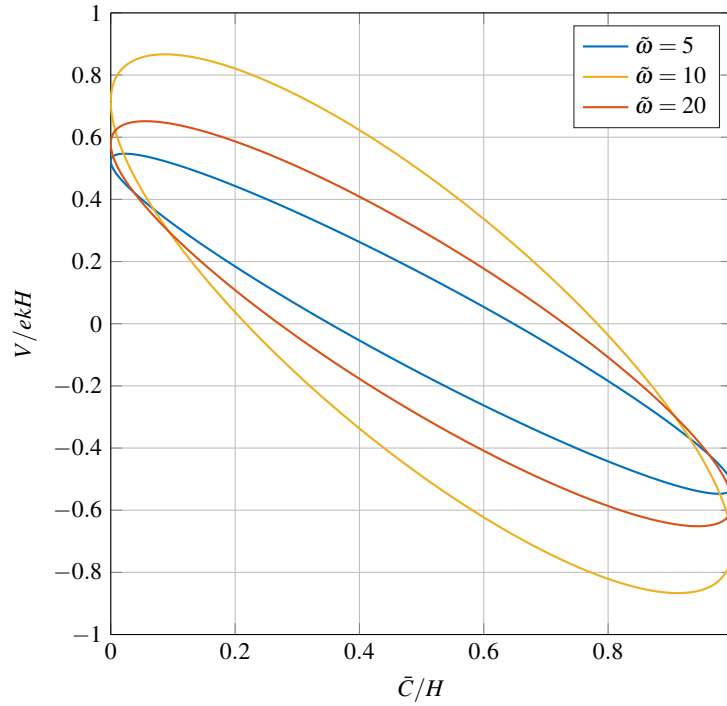
purely elasto-diffusive response only by constant fields depending on the initial state. Consequently, the corresponding charge-voltage curve in the cyclic steady state is directly obtained from the purely elasto-diffusive solution  $(c^E, \mu^E)$  (up to constants). Some example of such charge-voltage curves are shown in Fig. 4 for several values of the loading rate. The curves in Fig. 4 corresponds to the situation  $a\tilde{\omega} \ll k$ , in which case the chemical potential  $\mu^E(R, t)$  simplifies as  $\mu^E(R, t) = -\frac{Hk}{6}\Re(ze^{i\omega t})$ . Even though shakedown occurs and plastic dissipation vanishes in the cyclic steady state, the charge-voltage response in Fig. 4 shows some hysteresis (that increases with the charging rate). This results from the dissipation due to diffusion.

## CONCLUSION

Cyclically loaded media have been studied in the context of diffusion-induced plasticity. In the media considered, plasticity and diffusion act as two concurrent nonlinear and dissipative processes. Elastic shakedown corresponds to the situation in which plastic dissipation is bounded. Accordingly, the medium reaches a steady state cycle in which diffusion is the only active source of dissipation. For certain applications such as Li-ion batteries, elastic shakedown is of special interest both from mechanical and chemical considerations.

The presented Melan theorem gives a sufficient condition for elastic shakedown to occur, independently of the initial state. A general two-step procedure has been presented for applying that theorem in practice. The proposed two-step method allows one to estimate the set of loading parameters for which shakedown occurs – without resorting to step-by-step calculation of the chemo-mechanical evolution in elasto-plasticity. Interestingly, that two-step method allows for a decoupling between plasticity and diffusion. The first step is indeed completely disconnected from plasticity and consists in finding a cyclic steady-state  $(\sigma^E, \mu^E, u^E, c^E)$  for the purely elasto-diffusive problem. Once that elasto-diffusive problem is solved, the second step is performed without any explicit reference to diffusion as it only makes use of the stress  $\sigma^E$ . For the simple geometry considered, most of the needed calculations could be performed in closed-form. For more complex geometries, adequate numerical techniques would be necessary. In particular, Step 2 could be performed numerically by observing that it amounts to solve a convex programming problem, for which efficient algorithms (such as interior-point methods) are available.

In practice, Step 2. is performed by a considering a given set of self-equilibrated stress fields, resulting (in general) on lower bounds on the set of loading parameters for which elastic shakedown occurs. Upper bounds could possibly



**FIGURE 4.** Cyclic steady state for a spherical electrode particle in the elastic shakedown regime.

be achieved by extending the kinematic shakedown theorem of Koiter [4] to media coupling plasticity with diffusion.

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