



Chapter 9

Modelling of a Hydrogen Saturated Layer Within the Micropolar Approach

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Abstract This paper is concerned with modeling the strongly inhomogeneous hydrogen distribution over a sample by means of the micropolar continuum approach. The presence of micro-cracks covering the lateral surface of the sample is modeled by means of a distributed couple stress prescribed as a boundary condition. The applied couple stress produces a longitudinal displacement in return, which quickly fades away from the surface. The tensile displacement increases the intergranular space in the vicinity of the sample boundary and initiates hydrogen absorption from the environment. A comparison between widths of the surface layer that were experimentally determined and the ones that were analytically obtained allows estimating a value of one of the non-classical elastic parameters.

9.1 Introduction

Modern materials operate under extreme loads and in corrosive environments. The combined effect leads to stress corrosion cracking in metals (Jones, 2017), as well as to hydrogen-induced embrittlement (Koyama et al, 2012; Kyoung et al, 2009; Zhang et al, 2015). Experiments based on mass spectrometry and electron microscopy show that water vapor in the air is a source of hydrogen and that, in turn, the hydrogen leads to a decrease of strength, crack resistance and endurance lim-

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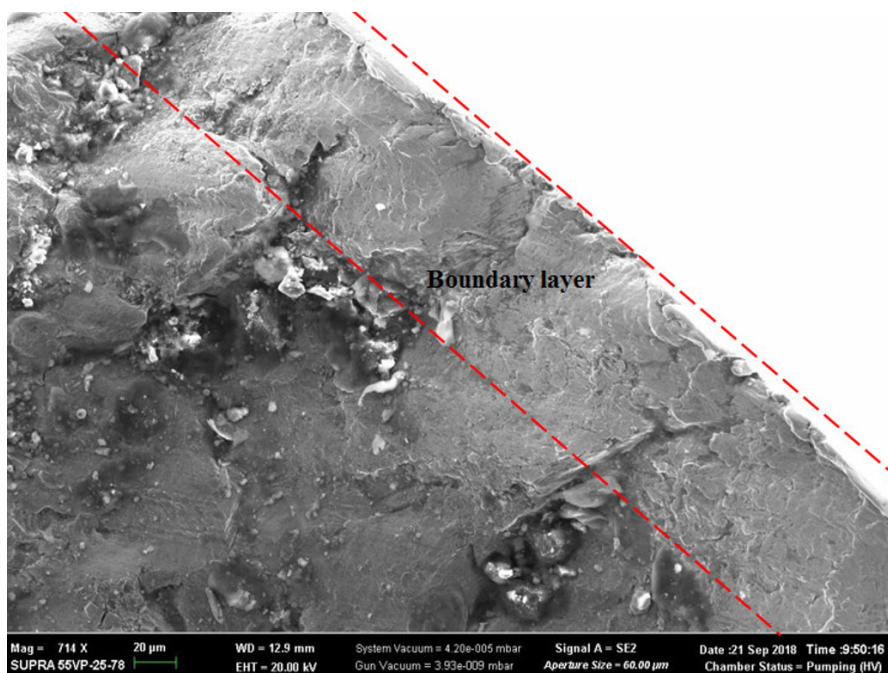


Fig. 9.1 Fractography of the sample after hydrogenation and fatigue test

its (Khrustalev et al, 1989). These effects are observed in samples saturated with hydrogen made from different types of materials, namely nickel, titanium, copper, aluminum alloys, and steel.

A number of articles were devoted to the investigation of hydrogen accumulation inside metals placed in an aggressive environment (Hadam and Zakroczymski, 2009; Martinsson and Sandström, 2012; Omura et al, 2016; Wu et al, 2015; Yagodzinsky et al, 2011). Direct measurements of hydrogen concentrations, as well as mathematical modeling of hydrogen accumulation, show that the hydrogen is unevenly distributed and that there is a significant excess of its concentration within a thin boundary layer of the metal (Martinsson and Sandström, 2012; Wu et al, 2015). A highly heterogeneous distribution of hydrogen within the samples is also observed when fatigue test are performed with non-hydrogen-charged samples placed in air environment (Belyaev et al, 2017a,b). In Fig. 9.1 the fractography of a sample made of alloy 718 after hydrogen charging and fatigue test is shown. In this case, the hydrogen concentration in a thin boundary layer with an approximate thickness of about 100 μ m exceeds 10–100 times the volumetric concentration.

It is known that the lateral surfaces of samples are covered with fractal oriented dislocations that, in turn, lead to micro-crack formation as in Betekhtin et al (2009); Kramer et al (2005); Steffens et al (1987). Detailed examinations of the surface layer in Steffens et al (1987) show that its grains or parts of a uniform mono-crystal rotate

and create empty spaces near the boundary. These additional empty spaces initiate the seepage of hydrogen from the external environment into the material. Thus, the non-homogeneous distribution of hydrogen over the sample can be explained by the existence of a lengthwise displacement appearing due to grain rotations in the vicinity of the lateral surface.

In other words, the theoretical interpretation of this phenomena should be connected with the material inner structure, in particular, with introduction of the intrinsic length scale parameter characterizing the width of the surface layer with a high hydrogen concentration. The dependence of mechanical response on the structure size could not be explained by the classical continuum mechanics since no length scale enters the constitutive equations. However, different generalized continuum theories have been successful in addressing a size effect problem. Examples for generalized continuum theories are the strain-gradient-theory as in Aifantis (1984); dell'Isola and Seppecher (1997); dell'Isola et al (2012), the micropolar theory as in Eringen and Kafadar (1976); Eremeyev et al (2012), and the surface-theory as in Altenbach et al (2010); Eremeyev and Pietraszkiewicz (2014), just to mention the most popular ones. A modern comparison of the different modelling options, which must be driven by phenomenology, can be found in dell'Isola et al (2017). In the present paper we are going to use a micropolar medium theory. As the first attempt to start the description and see how that approach works we will restrict ourself to isotropic linear continuum. A detailed discussion about other possible constitutive choices can be found in Eremeyev and Pietraszkiewicz (2016, 2012). Also it should be noted that although direct consideration of the cracking process is beyond the scope of the paper, the initial presence of the surface dislocations and microcracks is modeled by means of a distributed couple stress on the lateral surface of the sample. As a result, it will be shown that the prescription of couple stresses on the boundary leads to the appearance of additional tensile displacements increasing the intergranular space within the thin surface layer and therefore initiating the highly heterogeneous distribution of hydrogen.

Considering more complicated models usually involves introducing new parameters. There are four additional isotropic elastic constants in micropolar theory. Three of them provide a sensitivity to the rotation gradient and the remaining one quantifies the degree of coupling between macro and micro-rotation. Both theoretical and experimental investigations have been undertaken in order to determine these material coefficients (Adomeit, 1968; Askar, 1972; Askar and Cakmak, 1968; Ellis and Smith, 1967; Gauthier and Jahsman, 1975; Lake, 1983, 1995; Liebold and Müller, 2015, 2016; Perkins and Thompson, 1973; Schijve, 1966; Yang and Lakes, 1981). However, the accomplished work is still far from being complete and these coefficients are obtained only for a few different materials, namely, graphite, human bone, and foams. A comparison between the thickness of the high hydrogen concentration layer observed in experiments and the width of the layer with the analytically obtained tensile displacements allows for an estimate of the value of one of the non-classical elastic constants. This will be demonstrated in the last section for the aluminum alloy D16.

9.2 Basic Equations of Micropolar Media

Let us consider a geometrically linear micropolar continuum. Its deformations are described by the standard displacement field, \mathbf{u} , and an independent microrotation vector, $\boldsymbol{\theta}$. Then the stretch tensor \mathbf{e} and wryness tensor $\boldsymbol{\kappa}$ can be introduced in the following way¹ (Eremeyev et al, 2012):

$$\mathbf{e} = \nabla \otimes \mathbf{u} + \mathbf{I} \times \boldsymbol{\theta}, \quad (9.1)$$

$$\boldsymbol{\kappa} = \nabla \otimes \boldsymbol{\theta}, \quad (9.2)$$

where ∇ is the gradient operator, and \mathbf{I} is the unit tensor.

Note that the microrotation vector $\boldsymbol{\theta}$ is kinematically distinct from the “macro-rotation,” \mathbf{w} , determined by the antisymmetric part of the displacement gradient:

$$\mathbf{w} = \frac{1}{2} \nabla \times \mathbf{u}. \quad (9.3)$$

The vector $\boldsymbol{\theta}$ refers to the rotation of body particles, whereas \mathbf{w} refers to the rotation associated with translational motion of nearby body particles. Thus, the decomposition of the stretch tensor into symmetric and antisymmetric parts yields

$$\mathbf{e} = \boldsymbol{\epsilon} + \mathbf{I} \times (\boldsymbol{\theta} - \mathbf{w}), \quad (9.4)$$

where $\boldsymbol{\epsilon} = 1/2(\nabla \otimes \mathbf{u} + \mathbf{u} \otimes \nabla)$ is the strain tensor of classical linear elasticity.

The constitutive equations for the stress tensor \mathbf{T} and the couple stress tensor \mathbf{M} for linear isotropic Cosserat elasticity are as follows (Eringen and Kafadar, 1976):

$$\mathbf{T} = \lambda (\nabla \cdot \mathbf{u}) \mathbf{I} + 2\mu \boldsymbol{\epsilon} + \gamma \mathbf{I} \times (\boldsymbol{\theta} - \mathbf{w}), \quad (9.5)$$

$$\mathbf{M} = \beta_1 (\nabla \cdot \boldsymbol{\theta}) \mathbf{I} + \beta_2 \boldsymbol{\kappa}^\top + \beta_3 \boldsymbol{\kappa}, \quad (9.6)$$

where λ , μ , γ and β_i ($i = 1, 2, 3$) are independent elastic moduli. λ and μ are the classical elastic moduli, and γ is a modulus quantifying the degree of coupling between macro and micro rotation fields. The elastic moduli β_2 and β_3 allow to introduce material lengths reflecting effects of the couple stress, for example, $l_t = \sqrt{(\beta_2 + \beta_3)/2\mu}$ for torsion, or $l_b = \sqrt{\beta_3/4\mu}$ for bending (see, for example, Gauthier and Jahsman (1975)). When these length parameters vanish, the solutions obtained from couple stress theory reduce to those of classical elasticity theory. In perfect crystals and amorphous materials l_b and l_t are probably submicroscopic; but might be of the order of the averaged radius of roots of surface cracks (Mindlin, 1964).

The displacement and microrotation vectors can be found from the equilibrium equations, that in case of absence of body forces and body couples are

¹ A cross product between a second-rank tensor $\mathbf{A} = \mathbf{a}_k \mathbf{b}_k$ and vector \mathbf{c} is realized in the following way: $\mathbf{A} \times \mathbf{c} = \mathbf{a} \mathbf{b} \times \mathbf{c} = \mathbf{a} (\mathbf{b} \times \mathbf{c})$, i.e. the second vector in the tensor dyad is attached with the vector \mathbf{c} .

$$\nabla \cdot \mathbf{T} = 0, \quad (9.7)$$

$$\nabla \cdot \mathbf{M} + \mathbf{T}_\times = 0, \quad (9.8)$$

where $(\mathbf{a} \otimes \mathbf{b})_\times = \mathbf{a} \times \mathbf{b}$.

Within the framework of this paper we shall consider a case in which $\gamma \rightarrow \infty$. This corresponds to a case in which the antisymmetric part of the stretch tensor goes to zero or, in other words, to an assumption that the macrorotation and microrotation vectors are equal. It follows that it is the medium with constrained rotation, and the microrotation is determined by the displacement:

$$\boldsymbol{\theta} = \frac{1}{2} \nabla \times \mathbf{u}. \quad (9.9)$$

This means that $\nabla \cdot \boldsymbol{\theta} = 0$ and therefore parameter β_1 does not play any role in this model. Then Eqn.(9.6) simplifies to

$$\mathbf{M} = \frac{1}{2} (\beta_3 \nabla \otimes (\nabla \times \mathbf{u}) + \beta_2 (\nabla \times \mathbf{u}) \otimes \nabla). \quad (9.10)$$

According to Eqn. (9.8) together with Eqns. (9.5), (9.10), (9.4), and (9.3) we can obtain the following equality:

$$\mathbf{T}_\times = -2\gamma(\boldsymbol{\theta} - \mathbf{w}) = -\beta_3 \Delta \mathbf{w}, \quad (9.11)$$

where $\Delta = \nabla \cdot \nabla$ is the Laplace operator.

Consequently, the antisymmetric part of the stress tensor can be rewritten as follows:

$$\mathbf{T}^A = \frac{\beta_3}{4} \Delta (\mathbf{u} \otimes \nabla - \nabla \otimes \mathbf{u}). \quad (9.12)$$

As a result we obtain the following equation for the displacement field:

$$(\lambda + \mu) \nabla (\nabla \cdot \mathbf{u}) + \mu \Delta \mathbf{u} + \frac{\beta_3}{4} \nabla \cdot \Delta (\mathbf{u} \otimes \nabla - \nabla \otimes \mathbf{u}) = 0. \quad (9.13)$$

9.3 Axially-symmetrical Problem

In this paper we model the behavior of a cylindrical sample made of metal in an aggressive environment. In this context let us consider a boundary-value problem in cylindrical coordinates (r, φ, z) dealing with a solid cylinder of radius r_0 and length L subjected to a distributed couple stress, $-M_0 \mathbf{e}_\varphi$, on its lateral surface. Whereas a zero displacement field satisfies the equilibrium conditions and guarantees a traction-free lateral surface, the applied couple stress on the lateral surface produces an additional displacement which is fading away quickly from the surface.

We assume that the problem is axially symmetric and look for a solution with the following ansatz:

$$\mathbf{u} = \mathbf{u}(r, z) = u_r(r, z) \mathbf{e}_r + u_z(r, z) \mathbf{e}_z, \quad (9.14)$$

where \mathbf{e}_r is the radial unit vector, and \mathbf{e}_z is the unit vector along the cylinder axis.

From Eqns. (9.9) and (9.14) it follows that the microrotation vector has only one component, namely

$$\boldsymbol{\theta} = \theta_\varphi \mathbf{e}_\varphi = \frac{1}{2} \left(\frac{\partial u_r}{\partial z} - \frac{\partial u_z}{\partial r} \right) \mathbf{e}_\varphi. \quad (9.15)$$

In order to obtain the solution for the boundary layer in closed form, we have to take the following inequalities into account:

$$\begin{aligned} \frac{\partial^2 u_r}{\partial z^2} &\ll \frac{\partial^2 u_r}{\partial r^2}, \\ \frac{\partial^2 u_z}{\partial z^2} &\ll \frac{\partial^2 u_z}{\partial r^2}, \\ \frac{\partial u_z}{\partial z} &\ll \frac{\partial^2 u_z}{\partial r^2}. \end{aligned} \quad (9.16)$$

Consequently, Eqn. (9.13) reduces to

$$\begin{aligned} &\left[\lambda \left(\frac{\partial^2 u_r}{\partial r^2} + \frac{\partial^2 u_z}{\partial r \partial z} \right) + 2\mu \left(\frac{\partial^2 u_r}{\partial r^2} + \frac{1}{2} \frac{\partial^2 u_z}{\partial r \partial z} \right) - \frac{\beta_3}{2} \frac{\partial^3 \theta_\varphi}{\partial z \partial r^2} \right] \mathbf{e}_r + \\ &\left[\lambda \frac{\partial^2 u_r}{\partial z \partial r} + \mu \left(\frac{\partial^2 u_z}{\partial r^2} + \frac{\partial^2 u_r}{\partial r \partial z} \right) + \frac{\beta_3}{2} \frac{\partial^3 \theta_\varphi}{\partial r^3} \right] \mathbf{e}_z = 0. \end{aligned} \quad (9.17)$$

Here the terms of higher order of smallness are discarded.

By introducing non-dimensional parameters

$$\begin{aligned} x = 1 - \frac{r}{r_0}, \quad \tilde{z} = \frac{z}{L}, \quad \xi = \frac{r_0}{L}, \quad u_x = \frac{u_r}{r_0}, \quad u_{\tilde{z}} = \frac{u_z}{L}, \\ \tilde{\lambda} = \frac{\lambda}{\mu}, \quad \delta = \sqrt{\frac{\beta_3}{4\mu r_0^2}} = \frac{l_b}{r_0}. \end{aligned} \quad (9.18)$$

we can rewrite Eqn. (9.17) by the following system of equations:

$$\begin{aligned} &(\tilde{\lambda} + 2) \frac{\partial^2 u_x}{\partial x^2} - (\tilde{\lambda} + 1) \frac{\partial^2 u_{\tilde{z}}}{\partial x \partial \tilde{z}} - \delta^2 \frac{\partial^4 u_{\tilde{z}}}{\partial x^3 \partial \tilde{z}} = 0, \\ &-(\tilde{\lambda} + 1) \frac{\partial^2 u_x}{\partial x \partial \tilde{z}} + \frac{1}{\xi^2} \frac{\partial^2 u_{\tilde{z}}}{\partial x^2} - \delta^2 \left(\frac{1}{\xi^2} \frac{\partial^4 u_{\tilde{z}}}{\partial x^4} + \frac{\partial^4 u_x}{\partial x^3 \partial \tilde{z}} \right) = 0. \end{aligned} \quad (9.19)$$

By integrating the first equation, we obtain

$$\frac{\partial u_x}{\partial x} = \frac{\tilde{\lambda} + 1}{\tilde{\lambda} + 2} \frac{\partial u_{\tilde{z}}}{\partial \tilde{z}} + \frac{\delta^2}{\tilde{\lambda} + 2} \frac{\partial^3 u_{\tilde{z}}}{\partial x^2 \partial \tilde{z}}. \quad (9.20)$$

Then the second equation in (9.19) yields

$$u_{\tilde{z}} - \delta^2 \frac{\partial^2 u_{\tilde{z}}}{\partial x^2} = 0. \quad (9.21)$$

Since δ is of small value and the displacement has a finite value, we arrive at the following solution for $u_{\tilde{z}}$:

$$u_{\tilde{z}} = A(\tilde{z}) \exp\left(-\frac{x}{\delta}\right). \quad (9.22)$$

The solution for the dimensionless radial displacement reads

$$u_x = -A'(\tilde{z}) \delta \exp\left(-\frac{x}{\delta}\right). \quad (9.23)$$

By applying non-dimensional parameters, Eqn. (9.15) for the microrotations can be finally rewritten in the following way:

$$\theta_\varphi = \frac{1}{2\xi\delta} A(\tilde{z}) \exp\left(-\frac{x}{\delta}\right). \quad (9.24)$$

As a result the stress tensor has the form

$$\tilde{\mathbf{T}} = \frac{1}{\mu} \mathbf{T} = 2 \exp\left(-\frac{x}{\delta}\right) \left(A'(\tilde{z}) (\mathbf{e}_z \otimes \mathbf{e}_z - \mathbf{e}_r \otimes \mathbf{e}_r) + \frac{1}{\xi\delta} A(\tilde{z}) \mathbf{e}_z \otimes \mathbf{e}_r \right). \quad (9.25)$$

Obviously, the smallness of the first two terms is of higher order than the last one and formally they can be disregarded. However, in order to be sure that the lateral surface is traction free we will treat $A(z)$ as a constant. Thus the couple stress is given by

$$\tilde{\mathbf{M}} = \frac{1}{\mu L} \mathbf{M} = 2A \exp\left(-\frac{x}{\delta}\right) \left(\frac{\beta_2}{\beta_3} \mathbf{e}_\varphi \otimes \mathbf{e}_r - \mathbf{e}_r \otimes \mathbf{e}_\varphi \right). \quad (9.26)$$

From (9.26) we obtain the equation for the unknown constant:

$$\mathbf{e}_r \cdot \mathbf{M} |_{x=0} = -M_0 \mathbf{e}_\varphi, \quad \rightarrow \quad A = \frac{M_0}{2\mu L}. \quad (9.27)$$

So far we have considered a problem without external loads on the faces at the end. However, either axial tension

$$\mathbf{F} = F \mathbf{e}_z = \int_S T_{zz} \mathbf{e}_z \, dS$$

or torsion

$$\mathbf{M}_t = M_t \mathbf{e}_z = \int_S (\mathbf{r} \times (\mathbf{e}_z \cdot \mathbf{T}) + M_{zz} \mathbf{e}_z) \, dS$$

can be added. Here S is the cross-sectional area of the cylinder. The particular solutions of these Saint-Venant's problems are given by Gauthier and Jahsman (1975) and the total solutions for the medium with constrained rotation are

$$\begin{aligned}\mathbf{u} &= -\frac{\nu Fr}{ES}\mathbf{e}_r + \left(\frac{Fz}{ES} + \frac{M_0}{2\mu}\exp\left(-\frac{r_0-r}{l_b}\right)\right)\mathbf{e}_z, \\ \mathbf{T} &= \frac{F}{S}\mathbf{e}_z\mathbf{e}_z + \frac{M_0}{l_b}\exp\left(-\frac{r_0-r}{l_b}\right)\mathbf{e}_z\mathbf{e}_r\end{aligned}\quad (9.28)$$

for tension and

$$\begin{aligned}\mathbf{u} &= C_1 r z \mathbf{e}_\varphi + \frac{M_0}{2\mu}\exp\left(-\frac{r_0-r}{l_b}\right)\mathbf{e}_z, \\ \mathbf{T} &= C_1 \mu r (\mathbf{e}_z \otimes \mathbf{e}_\varphi + \mathbf{e}_\varphi \otimes \mathbf{e}_z) + \frac{M_0}{l_b}\exp\left(-\frac{r_0-r}{l_b}\right)\mathbf{e}_z \otimes \mathbf{e}_r, \\ \mathbf{M} &= C_1 (\beta_2 + \beta_3) \left(\mathbf{e}_z \otimes \mathbf{e}_z - \frac{1}{2}(\mathbf{e}_r \otimes \mathbf{e}_r + \mathbf{e}_\varphi \otimes \mathbf{e}_\varphi)\right) + \\ &+ M_0 \exp\left(-\frac{r_0-r}{l_b}\right) \left(\frac{\beta_2}{\beta_3}\mathbf{e}_\varphi \otimes \mathbf{e}_r - \mathbf{e}_r \otimes \mathbf{e}_\varphi\right), \\ C_1 &= \frac{M_t}{S} \left(\beta_2 + \beta_3 + \mu \frac{r_0^2}{2}\right)^{-1}\end{aligned}\quad (9.29)$$

for torsion. Here $\nu = \lambda/(2(\lambda + \mu))$ and $E = \mu(3\lambda + 2\mu)/(\lambda + \mu)$ are Poisson's ratio and Young's modulus, respectively.

In both cases, the additional exponential terms play a role only within a thin surface layer and serves as a correction terms in order to satisfy the boundary condition on the lateral surface. Note that the torsion solution provided in Gauthier and Jahsman (1975) is traction-free on the lateral surface. However, in the case of the medium with constrained rotation it has a couple stress on the boundary:

$$M_{rr} = -\frac{C_1}{2}(\beta_2 + \beta_3). \quad (9.30)$$

9.4 Results

As one can see from the previous section, considering a micropolar media allows us to obtain an additional displacement along the cylinder axis in the vicinity of the lateral surface. This tensile displacement increases the intergranular space and can initiate hydrogen absorption from the environment. In order to estimate the parameters of the model let us consider a cylindrical sample made of the aluminum alloy D16, because this material is frequently used in experiments in Andronov et al (2017). The following values will be used: $r_0 = 0.0045$ m, $L = 0.035$ m, $\mu = 27000$ MPa.

In order to estimate the small parameter δ we correlate the width of the layer with the additional displacements within the thickness of the boundary layer containing a higher concentration of hydrogen, x_* . Since u_z is exponentially decreasing from of the surface we define its limit value by $\chi u_z|_{x=0}$, where χ is a small parameter. Thus, the equation for δ can be written in the following way:

$$\delta = -\frac{x_*}{Ln(\chi)}. \tag{9.31}$$

When taking $x_* = 100 \mu\text{m}/r_0 \approx 0.02$ and $\chi = 0.01$ we obtain $\delta \approx 0.005$. Recall that δ is related to the non-classic elastic modulus β_3 through Eqn. (9.18) as follows:

$$\beta_3 = 4\mu r_0^2 \delta^2. \tag{9.32}$$

Therefore, the elastic modulus can be estimated as: $\beta_3 \approx 5 \cdot 10^{-5} \text{ MPa m}^2$.

The dependence of the displacement along the cylinder axis for this value of β_3 is shown in Fig. 9.2. The curve is plotted for $M_0 = -189 \text{ kPa}$ corresponding to $A = 0.0001$.

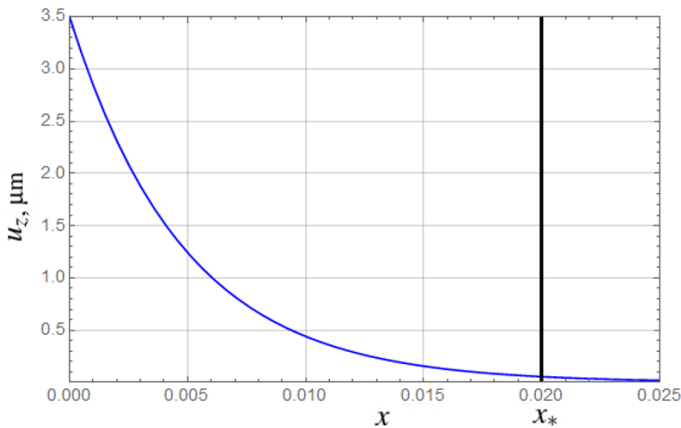


Fig. 9.2 Dependence of the tensile displacement on the relative distance from the lateral surface

9.5 Conclusions and Outlook

In this paper two main tasks were accomplished:

- First, an attempt was made to model the strongly inhomogeneous distribution of hydrogen observed in experiments within a micropolar continuum approach. The stress-strain state in the sample can be obtained within the theory of classical elasticity while the situation near the body surface could be depicted more

accurately by a solution, which is quickly fading away from the border. This solution gives additional tensile displacements in the vicinity of the lateral surface by means of which the absorption of hydrogen from the environment can be initiated. Note that we did not consider the hydrogen diffusion but concentrated on possible reasons of the hydrogen seepage.

- Second, a new method for estimating the additional elastic constants of a Cosserat medium was proposed. It is based on the comparison between the experimentally measured width of the surface layer containing the high hydrogen concentration and the analytically obtained characteristic length from the fading solution.

More research in this field is planned for the future in order to study the boundary layer emergence within Cosserat elasticity without restriction. At this stage a medium with constrained rotation was considered. The adopted simplification allowed us to obtain the analytical solution and to demonstrate feasibility of the suggested approach. However, the strong coupling between the macro and micro-rotations does not describe the rotation of grains in a proper manner and should be softened.

Moreover, if the displacement was supposed to be axially symmetric. This is a “classical” assumption for a cylindrical sample, but it limits the types of boundary condition to be prescribed at the lateral surface, namely, the distributed couple stress caused by microcracks. The microcracks distribution might be non-homogeneously over the sample surface and therefore leads to a dependence of the couple stress on the azimuth angle φ . Evidently a numerical approach will be required in that case.

In order to estimate the value of the non-classical elastic parameters, further efforts should be devoted to providing experiments with different materials and specimens sizes.

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