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A COUPLED THEORY OF SOLUTE DIFFUSION AND DEFORMATION IN ELASTIC SOLIDS

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Abstract. *This paper deals with a continuum theory for the interaction between solute diffusion and deformation in elastic solids. A salient feature of this continuum model relies on the conception of the solid-solute mixture as a solid with a microstructure through which the solute can move independently. Further, the solute can distort the solid, which otherwise is conserved. Specifically, within a small strain and isothermal setting, the purpose of this work is twofold: a) to use modern continuum mechanics to develop a framework for the description of the interaction of solute diffusion and deformation in elastic solids; b) to specialize the general framework to obtain a simple set of governing equations which demonstrate that the general framework is capable of capturing some of the most essential features of the interplay between diffusion and deformation. As an application, we consider the problem of diffusion-induced bending in beams. This issue is of particular relevance for the understanding of microcantilever sensors, which have been used with success for the detection of small amounts of solute molecules, hydrogen molecules for instance, in air and solution.*

Keywords: *diffusion, elastic solids, solute transport, diffusion-induced bending, hydrogen sensors*

1. INTRODUCTION

Studies of mechanical phenomena associated with changes in the chemical environment have a substantial historical background. In 1924, Palmer (1924) studied coherence of loosely contacting thin filaments induced by electromagnetic waves in the presence of different gases and analyzed correlation between the observed responses and the heat of gas absorption. Almost at the same time, Mehaan (1927) observed adsorption-induced expansion of yellow pine charcoal exposed to carbon dioxide vapors and showed that this was a reversible process. Later, Yates (1954) conducted similar studies on the expansion of porous glass exposed to nonpolar gases such as Ar, N_2 , O_2 , H_2 , and Kr.

Mechanical stresses and deformations produced in response to a changing chemical environment have also drawn attention as a principle of powering miniature mechanical devices. For example, in the 1960s, Kuhn *et al.* (1950) and Steinberg *et al.* (1966) developed the concept of devices that provide direct conversion of chemical stimuli into mechanical energy. For example, in Sussman and Katchalsky (1970), such devices referred as mechanochemical engines has not been pursued since then primarily because of the difficulty associated with microfabrication. Furthermore, practical implications of these devices were limited until advances in microtechnology and, more recently, in MEMS opened up an opportunity to fabricate miniaturized mechanical components routinely

This paper presents briefly a continuum theory for the description of the interaction between solute diffusion and deformation in elastic solids, developed by Duda et al (2009a,b). In this theory we describe the behaviour of a solid undergoing two interdependent processes, a macroscopic or mechanical process due to the deformation of the solid, and a microscopic or chemical process due to the migration of a species through the solid. In accordance with the formalism of continuum mechanics we consider the following topics: basic balances; free energy imbalance (the second law of thermodynamics) and constitutive theory.

The free energy imbalance accounts for energy inflow due to both power expended by all external forces and solute supply, a contribution through which the chemical potential is introduced. The constitutive theory is based upon the following assumptions: additive decomposition of the solid strain into its elastic and solute-induced parts; the dissipation is due diffusion; the set of constitutive variables includes elastic strain and solute density. Finally, the underlying governing equations of the framework are obtained by merging the aforementioned ingredients.

As an application, we consider the problem of diffusion-induced bending in beams. This issue is of particular relevance for the understanding of microcantilever sensors, which have been used with success for the detection of small amounts of solute molecules, hydrogen molecules for instance, in air and solution.

A hydrogen cantilever microsensor detects the presence of low concentration of hydrogen in a small volume and measures the surface stress associated with the molecular adsorption or absorption. The data acquisition and interpretation are based on the deflection of cantilevers. The diffusion induced stress and deformation in microcantilever sensors and actuators has been studied recently. Based on the theory of linear elasticity Yang and Li (2003) analyzed the diffusion-

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induced beam bending in hydrogen sensors without consideration of mechanical loads. Xuan *et al.* (2010) analyzed the influence of residual on diffusion-induced bending in bilayered microcantilever sensors, but also without consider the mechanical loads.

The purpose of this article is to analyze the bending of a beam due a diffusion of hydrogen molecules and mechanical loads.

2. The continuum model

In this section we briefly present the coupled model for deformation and solute diffusion in solids.

Consider a solid $\boldsymbol{\mathcal{B}}$ capable of absorbing a fluid-like chemical species, or simply a solute for the sake of brevity. There are two processes taking place at two different scales, a macroscopic (mechanical) process due to the deformation of the solid and a microscopic (chemical) process due to the migration of the solute through the solid. The state of $\boldsymbol{\mathcal{B}}$ at time t is defined by the vector field $\mathbf{u}$ representing the mechanical motion (the displacement),and the scalar field α representing the fluid motion. The corresponding realizable velocities are $\mathbf{v} = \dot{\mathbf{u}}$ and $\mu = \dot{\alpha}$, interpreted as the chemical potential. We denote by c and c_s the solute and the solid content per unit reference volume, respectively, and by ϕ the solute concentration.

$$\phi = \frac{c}{c_s} \tag{1}$$

We present the mechanical and chemical balances, following the approach by Duda *et al.* (2009b), in the setting of the infinitesimal theory. For any part of $\boldsymbol{\mathcal{P}}$ of $\boldsymbol{\mathcal{B}}$ we have

$$\begin{cases} \operatorname{Div}\mathbf{T} + \mathbf{b} = \mathbf{0} \quad \text{and} \quad -\operatorname{Div}\mathbf{J} + h = \dot{c} \quad \text{on } \boldsymbol{\mathcal{P}}, \\ \mathbf{T}\mathbf{n} = \mathbf{t} \quad \text{and} \quad -\mathbf{J}\cdot\mathbf{n} = j \quad \text{on } \partial\boldsymbol{\mathcal{P}}, \end{cases} \tag{2}$$

where $\mathbf{T}$ is the Cauchy stress tensor, $\mathbf{b}$ is the body force vector, $\mathbf{J}$ is the referential flux of the solute relative to the solid, h is the solute supply, and $\mathbf{n}$ is the exterior unit normal to $\partial\boldsymbol{\mathcal{P}}$.

The dissipation inequality is given by

$$\dot{\psi} - \mathbf{T}\cdot\dot{\mathbf{E}} + \mathbf{J}\cdot\nabla\mu - \dot{c}\mu \leq 0, \tag{3}$$

where ψ is the free-energy density and $\mathbf{E}$ is the infinitesimal strain tensor, given by

$$\mathbf{E} = 1/2(\nabla\mathbf{u} + \nabla\mathbf{u}^T) := (\nabla\mathbf{u})^S. \tag{4}$$

We assume the additive decomposition of the strain tensor $\mathbf{E}$ into its elastic $\mathbf{E}_e$ and solute induced $\mathbf{E}_s$ parts

$$\mathbf{E} = \mathbf{E}_e + \mathbf{E}_s \qquad \text{with} \qquad \mathbf{E}_s = \eta\,(\phi - \phi_0)\mathbf{I}, \tag{5}$$

where η is a positive parameter and ϕ_0 a reference solute concentration. It follows then that the dissipation inequality (3) can be written as

$$\dot{\psi} - (\mu + \frac{\eta}{c_s}\operatorname{tr}\mathbf{T})\,\dot{c} - \mathbf{T}\cdot\dot{\mathbf{E}}_e + \mathbf{J}\cdot\nabla\mu \leq 0. \tag{6}$$

Based on the inequality (6), we conclude that

$$\psi = \hat{\psi}(\mathbf{E}_e, c), \quad \mathbf{T} = \hat{\mathbf{T}}(\mathbf{E}_e, c) = \frac{\partial\hat{\psi}(\mathbf{E}_e, c)}{\partial\mathbf{E}_e}, \quad \mu + \frac{\eta}{c_s}\operatorname{tr}\mathbf{T} = \frac{\partial\hat{\psi}}{\partial c}(\mathbf{E}_e, c), \quad \hat{\mathbf{J}}(\mathbf{E}_e, c, \nabla\mu)\cdot\nabla\mu \leq 0, \tag{7}$$

it can be shown that $(7)_4$ implies the representation

$$\hat{\mathbf{J}}(\mathbf{E}_e, c, \nabla\mu) = -\hat{\mathbf{M}}(\mathbf{E}_e, c, \nabla\mu)\nabla\mu, \tag{8}$$

where $\mathbf{M}$ is a positive semidefinite tensor-valued function related to the solute mobility.

2.1 Specialization

We assume that the free-energy density $\hat{\psi}(\mathbf{E}_e, c)$ is given by

$$\hat{\psi}(\mathbf{E}_e, c) = \frac{\bar{\lambda}}{2}(\operatorname{tr}\mathbf{E}_e)^2 + \bar{\mu}|\mathbf{E}_e|^2 + k_B T c\left[\left(\ln\frac{c}{c_s}\right) - 1\right], \tag{9}$$

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where the first two terms correspond to the classical elastic strain energy, with $\bar{\lambda}$ and $\bar{\mu}$ Lamé parameters. The last term represents the classical entropic contribution to the free energy of a dilute ideal interstitial solid solution, where k_B is Boltzmann's constant and T is the absolute temperature. We also assume that the tensor $\hat{\mathbf{M}}$ in Equation (8) is given by:

$$\hat{\mathbf{M}}(\mathbf{E}_e, c, \nabla\mu) = Mc\mathbf{I} \quad \text{and then} \quad \mathbf{J} = -Mc\nabla\mu, \tag{10}$$

where M is a positive parameter

2.2 Governing equations

The governing equations for the fields $\mathbf{u}$, μ, $\mathbf{J}$ and c are obtained through the combination of the aforementioned basic balances and constitutive relations. Body force, inertial effects, and solute supply are neglected here. The corresponding equations can be written as

$$\begin{aligned} &\mathrm{Div}\mathbf{T} = \mathbf{0}, \\ &-\mathrm{Div}\,\mathbf{J} = \dot{c}, \end{aligned} \tag{11}$$

where:

$$\begin{aligned} &\mathbf{T} = \bar{\lambda}\,(\mathrm{tr}\mathbf{E}_e)\mathbf{I} + 2\bar{\mu}\mathbf{E}_e, \qquad \mathbf{E}_e = (\nabla\mathbf{u})^S - \eta\,(\phi - \phi_0)\mathbf{I}, \qquad \phi = \frac{c}{c_s}, \\ &\mu = k_B T \ln\phi - \frac{\eta}{c_s}\,\mathrm{tr}\,\mathbf{T}, \\ &\mathbf{J} = -\frac{D}{k_B T}\,c\,\nabla\mu, \quad D = Mk_B T, \end{aligned} \tag{12}$$

The equations $(11)_1$ and $(11)_2$ are the local form of the balance equation for the momentum (see $(2)_1$) and the balance equation for the solute content (see $(2)_2$), respectively. Equations $(12)_1$ is the equilibrium equation (see $(7)_5$), we can observe that if $\mathbf{E}_e = \mathbf{0}$, then there are no stress in the system. Equation $(12)_2$ represents the additive decomposition of the strain tensor (see (5)). The equations $(12)_3$-$(12)_5$ correspond to the microscopic process due to the migration of the solute through the solid. The equation $(12)_4$ provides a generalization of Fick's law since the solute flux may be driven by the solute content, elastic strain and chemical potential gradients (see (10)), where D is the diffusion coefficient. Equation $(12)_3$ is the constitutive equation (see $(7)_4$).

3. Diffusion-induced beam bending

The theory presented in this section is similar the theory of thermal stresses described by Boley and Weiner (1997). We consider a beam exposed to a diffusing component and free of external (mechanical) loads. We denote the axial coordinate by x, and letting y and z be centroidal axes in the cross-sectional planes. The diffusion-induced bending, in the plane $x - y$ or $x - z$, can be evaluated under the Euler-Bernoulli assumption that sections which are plane and perpendicular to the axis before loading remains so after loading. Therefore, we consider the axial stress T_{xx} and the axial strain E_{xx}. The axial displacement can be written as

$$u(x, y, z) = f_0(x) + yf_1(x) + zf_2(x). \tag{13}$$

The corresponding axial normal strain and stress components are then (see equations (4), (5), $(12)_1$ and $(12)_2$)

$$\begin{aligned} &E_{xx} = \frac{\partial u}{\partial x} = f_0'(x) + yf_1'(x) + zf_2'(x), \\ &T_{xx} = E\,(E_{xx} - \eta\,(\phi - \phi_0)), \end{aligned} \tag{14}$$

where E is Young's modulus of the material. The functions f_0, f_1 and f_2 must be determined so as satisfy the equations of equilibrium, calculated across the cross section

$$\int_A T_{xx}\,\mathrm{dA} = \int_A T_{xx}\,y\,\mathrm{dA} = \int_A T_{xx}\,z\,\mathrm{dA} = 0, \tag{15}$$

where the integrations must be extended over the cross-sectional area A. Considering that x, y, z are principal axes, we have

$$\int_A y\,\mathrm{dA} = \int_A z\,\mathrm{dA} = \int_A yz\,\mathrm{dA} = 0 \qquad \text{and} \qquad \int_A \mathrm{dA} = A, \quad \int_A y^2\,\mathrm{dA} = I_z, \quad \int_A z^2\,\mathrm{dA} = I_y, \tag{16}$$

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where I_y and I_z are the moments of inertia about y and z axes, respectively. Substituting equation $(14)_2$ into equations (15), and considering the relations described in (16), we have:

$$f_0' = \frac{\eta}{A}\int_A \phi\,\mathrm{dA}, \quad f_1' = \frac{\eta}{I_z}\int_A \phi\, y\,\mathrm{dA} \quad \text{and} \quad f_2' = \frac{\eta}{I_y}\int_A \phi\, z\,\mathrm{dA}, \tag{17}$$

Substituting equations (14) into equations (15), we have the final expression for T_{xx}

$$\begin{aligned} T_{xx} &= \frac{M_{cz}\,y}{I_z} + \frac{M_{cy}\,z}{I_y} - E\eta(\phi - \phi_0 - \bar{\phi}), \\ \text{where} \quad \bar{\phi} &= \frac{1}{A}\int_A \phi\,\mathrm{dA}, \quad M_{cy} = E\eta\int_A \phi\, z\,\mathrm{dA}, \quad M_{cz} = E\eta\int_A \phi\, y\,\mathrm{dA}, \end{aligned} \tag{18}$$

and, substituting equations (17) into equation $(14)_1$, and considering the relations defined in $(18)_{2-4}$, we have:

$$E_{xx} = \frac{M_{cz}\,y}{EI_z} + \frac{M_{cy}\,z}{EI_y} + \eta\,\bar{\phi}. \tag{19}$$

The radius of curvature ρ of the centroidal plane, in the plane $x-y$ or $x-z$ is

$$\frac{1}{\rho_y} = \frac{\partial E_{xx}}{\partial y} = \frac{M_{cz}}{EI_z} \qquad \text{or} \qquad \frac{1}{\rho_z} = \frac{\partial E_{xx}}{\partial z} = \frac{M_{cy}}{EI_y} \tag{20}$$

Similar expression is obtained in mechanical bending under the Bernoulli-Euler assumption, that is, $1/\rho = M/EI$. Therefore we conclude

$$\frac{\partial^2 v}{\partial x^2} = \frac{M_{cz}}{EI_z} \qquad \text{or} \qquad \frac{\partial^2 w}{\partial x^2} = \frac{M_{cy}}{EI_y}, \tag{21}$$

where $v(x,y,z)$ and $w(x,y,z)$ are the deflections in the y and z directions respectively.

An additional curvature term must be included in equations (21) if transverse loads are present

$$\frac{\partial^2 v}{\partial x^2} = \frac{M_{cz} + M_z}{EI_z} \qquad \text{or} \qquad \frac{\partial^2 w}{\partial x^2} = \frac{M_{cy} + M_y}{EI_y}, \tag{22}$$

where M_y and M_z are the bending moments due mechanical loads, that cause rotation about the y or z axes, respectively

4. Application

Consider a beam of thickness $2h$ in the y direction and width b in the z direction, and suppose that $M_{cy} = 0$, that is, the bending moments cause rotation about the z axe. In this case $v = v(x)$ and $w = 0$. From Equations (18) and (22), we have

$$\begin{aligned} T_{xx} &= \frac{M_{cz}\,y}{I_z} - E\eta(\phi - \phi_0 - \bar{\phi}) \quad \text{where} \quad \bar{\phi} = \frac{1}{2h}\int_{-h}^{h} \phi(y)\,\mathrm{dA}, \quad M_{cz} = E\eta b\int_{-h}^{h} \phi(y)\,y\,\mathrm{dA}, \\ \frac{\mathrm{d}^2 v}{\mathrm{d}x^2} &= \frac{M_{cz} + M_z}{EI_z}. \end{aligned} \tag{23}$$

Consider the beam also subjected to an external force per unit length $f(x)$. From the theory of bending of beams, we have:

$$\frac{\mathrm{d}^2 M_z}{\mathrm{d}x^2} = f(x). \tag{24}$$

From equations $(23)_4$ and (24)

$$\frac{\mathrm{d}^2}{\mathrm{d}x^2}\left(EI_z\frac{\mathrm{d}^2 v(x)}{\mathrm{d}x^2} - M_{cz}\right) = f(x). \tag{25}$$

The equation (25) in the weak form is

$$\int_0^L \left(EI_z\frac{d^2 v}{dx^2} - M_{cz}\right)\frac{d^2\eta}{dx^2}dx = \int_0^L f\eta dx + \left(EI_z\frac{d^2 v}{dx^2} - M_{cz}\right)\frac{d\eta}{dx}\bigg|_0^L - \frac{d}{dx}\left(EI_z\frac{d^2 v}{dx^2} - M_{cz}\right)\eta\bigg|_0^L \tag{26}$$

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Observe that, in this model there is a coupling effect in the equation that describes the chemical potential (see equation $(12)_3$), i.e., coupling between c and $\mathbf{T}$. The model described by Yang and Li (2003) (a simpler one) does not consider the stress effect. Therefore, we do not consider the stress term in that equation. Hence, in one dimensional setting, the diffusion Equation (see equations $(11)_2$ and $(12)_{3,4,5}$) is written as

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial y^2} \tag{27}$$

Now consider a beam with one surface exposed to a diffusing solute which maintains a concentration fixed in c_h and the other surface is impermeable. At the instant $t = 0$ there is no such component in the beam. Therefore we have the following boundary conditions

$$\begin{aligned} &c(y) = 0 \quad \text{at} \quad t = 0, \\ &c\,(h) = c_h \quad \text{and} \quad \left.\frac{\partial c}{\partial y}\right|_{y=-h} = 0 \quad \text{for} \quad t > 0. \end{aligned} \tag{28}$$

Equation for the concentration at any time t for any y is (see Lee *et al.* (2000))

$$\frac{c}{c_h} = 1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n+1)}\cos\left[\left(\frac{2n+1}{4h}\pi\right)(y+h)\right] \times \exp\left[-\left(\frac{2n+1}{4h}\pi\right)^2 Dt\right] \tag{29}$$

The equation that describes M_{cz} (see $(23)_3$) can be written as (see Lee *et al.* (2000))

$$M_{cz}(t) = -\frac{16bh^2c_h}{\pi^3}\sum_{n=0}^{\infty}\frac{(2n+1)\,\pi - 4\,(-1)^n}{(2n+1)^3} \times \exp\left(-\frac{(2n+1)^2\,\pi^2 Dt}{16h^2}\right) \tag{30}$$

5. Numerical example

In this section solutions to the initial boundary value problem of transient hydrogen diffusion coupled with elasticity are presented in a single-layer high strenght steel cantilever. The units used are: the solid density c_s measured in host lattice atoms (Fe) per volume, i.e., (Fe/m^3), and the solute density c measured in hydrogen atoms (H) per volume, i.e., (H/m^3). The solute concentration $\phi = c/c_s$, is, then, measured in hydrogen atoms (H) per host lattice atoms (Fe), i.e., H/Fe. And the cantilever dimensions are: a lenght of $l = 10$ mm, a thickness of $h = 0.25$ mm and a width of $b = 1.5$ mm. For the diffusion boundary and initial conditions we assume that one surface exposed to difussing component maintains a fixed concentration of $\phi = 8.5\text{x}10^-3$ H/Fe while the opposite surface is impermeable and that at $t = 0$ the cantilever has no concentration of the component. The material properties used herein were the same adopted by Duda and Souza (2009a).

Table 1. Material properties of high strenght steel with internal hydrogen

λ	119.0 GPa
$\bar{\mu}$	79.0 GPa
D	1.0x10^{-8} m^2/s
c_s	8.454x10^{28} Fe/m^3
k_BT	4.14x10^{-21} Joule
η	0.9307 Fe/H

The solute concentration distributions with different times of the free of external loading cantilever are plotted in Fig.1. The solid line corresponds to initial instant, when the difussion process had barely started. The solute difuses from the negative side surface of the cantilever into the positive side surface, the process go on increasing with time, until the saturation.

As shown in Fig.2 the diffusive moment M_{cz} reaches its maximum value after the firsts hydrogen atoms diffuse. This is because, at the early stage of the diffusion, the gradient generated in the axial deformation due to the presence of these atoms is maximum. As the time increases the diffusive moment goes to zero.

As prediced by the diffusive moment, the cantilever deflection has the maximum value at the early stage of the diffusion, and by the end of the diffusion process there will be no deflection as shown in Fig.3. Figure 4 compares the deflection induced by the diffusive moment at 1 second with and without an uniform external mechanical load on negative y direction.

The corresponding axial normal stress and strain component for different times are plotted in Figures 5 and 6 respectively.

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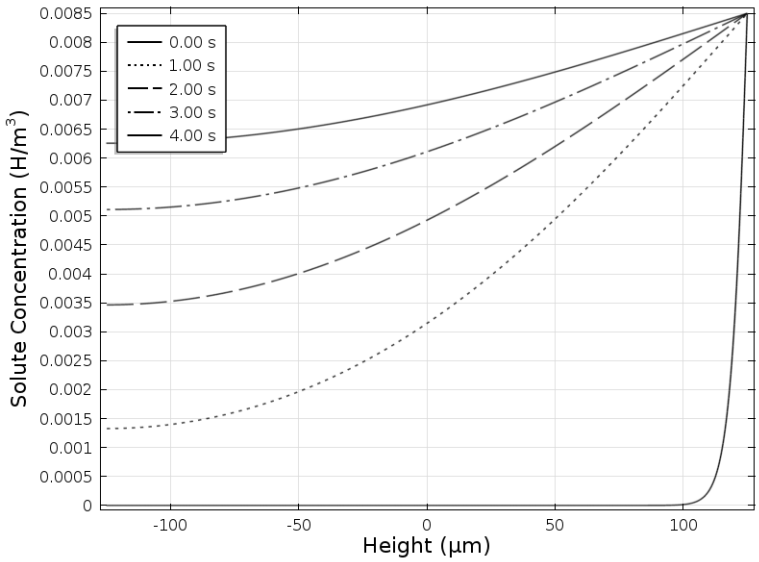

Figure 1. Solute distribution

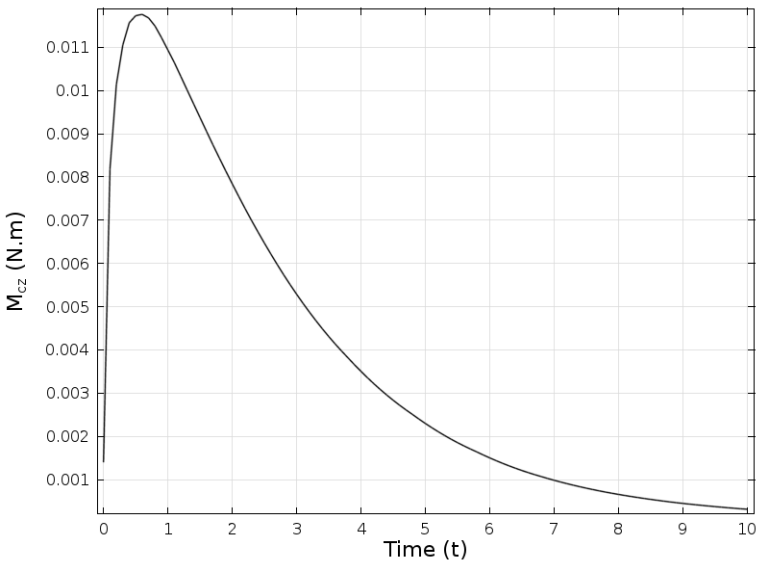

Figure 2. Diffusive moment

6. ACKNOWLEDGEMENTS

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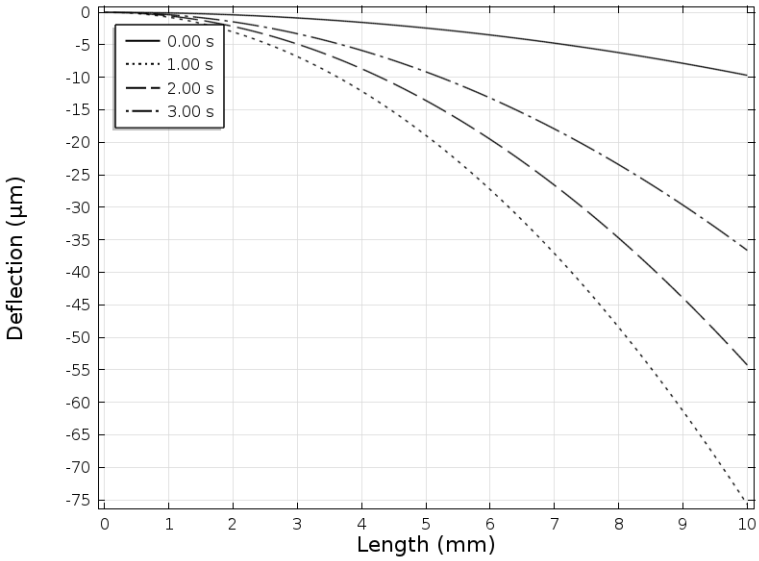

Figure 3. Beam deflection

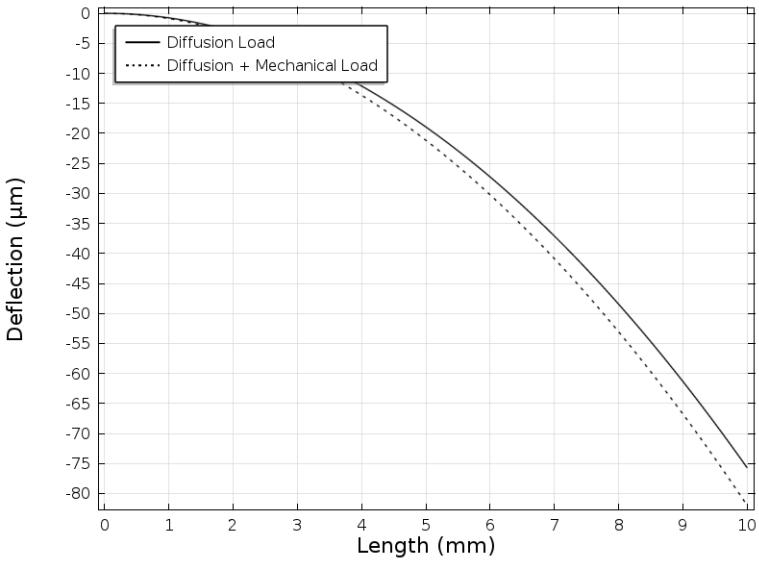

Figure 4. Mechanical loading comparison

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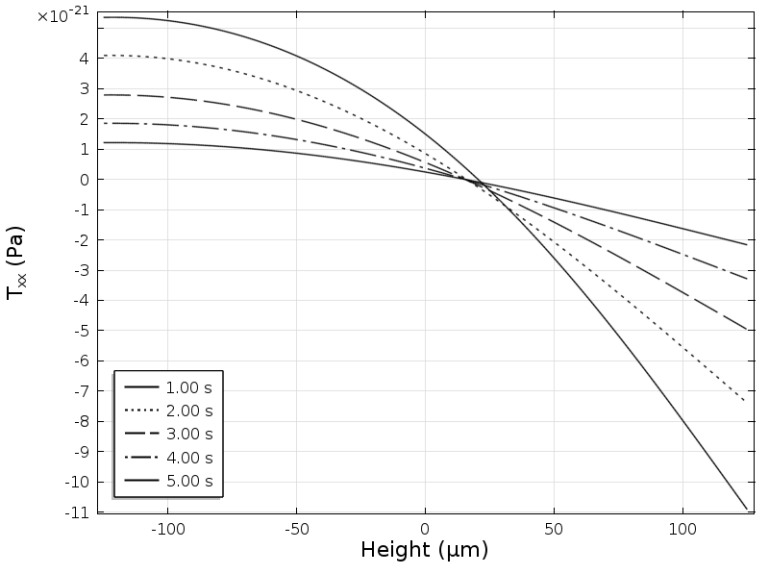

Figure 5. T_{xx} along the diffusion direction

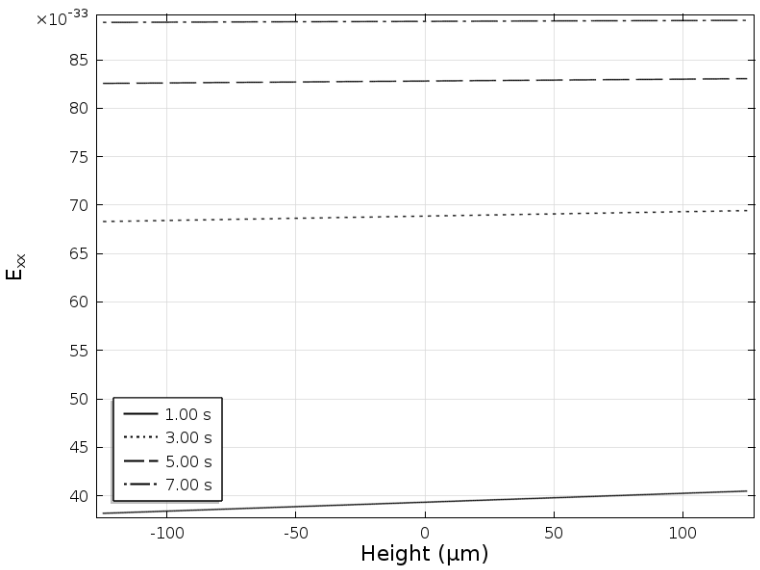

Figure 6. E_{xx} along the diffusion direction