Chapter 11
Wave Nature of Hydrogen Concentration Dynamics in Materials

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Abstract The models of the wave description of the hydrogen concentration in materials are discussed and compared. Special attention is paid to the role of nonlinear effects.

Keywords Hydrogen concentration · Bi-continuum · Nonlinear effects
Nonlinear wave modeling

11.1 Introduction

The influence of hydrogen on the plasticity and strength of metals attracts considerable attention in the last years. One can say that the main problem today is hydrogen-induced destruction. The modeling of hydrogen dynamics concerns various approaches. One of them considers the evolution of the hydrogen concentration as a wave process.

The wave description was less developed recently because of the linear description. The hydrogen transfer has been modeled by the equations of diffusion type. However, the linear diffuison equation doesn't possess wave solutions with finite velocity. Only recently, such models began to appear [1–3]. Later, the wave character of concentration has been detected in experiments [3–5]. The model developed in [2] has been extended and thoroughly examined in [5–7]. Also, an influence of the strains in the material on the concentration dynamics has been modeled in [8] on the basis of discrete-continuum nonlinear modeling. All these studies allowed to reveal new phenomena described with the help of a nonlinear description.

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The aim of the paper is to call attention to the nonlinear wave modeling of the hydrogen concentration and the sources of nonlinearity causing qualitatively new features of the concentration dynamics.

11.2 Bi-Continuum Nonlinear Model

This model has been developed in [2, 3, 6, 7].

Assume the diffuse hydrogen is characterized by m_H, v_H^- and ρ_H^- which are the mass, velocity and volume density respectively. Similarly, the bound hydrogen is described by v_H^+, and ρ_H^+.Then the concentrations are $N_H^- = \rho_H^-/m_H, N_H^+ = \rho_H^+/m_H$.

Then basic equations in the one-dimensional case are

$$(\rho_0 + m_H\, N_H^+)\, v_{H,t}^+ + (F_0 - \gamma N_H^+) m_H\, N_H^-(v_H^- - v_H^+) + (\alpha\, N_H^- - \beta\, N_H^+)\, v_H^+ = \sigma_x, \tag{11.1}$$

$$m_H\, N_H^-\, v_{H,t}^- + (F_0 - \gamma N_H^+) m_H\, N_H^- v_H^- + \frac{3}{2} k\, T\, N_{H,x}^- + (\alpha\, N_H^- - \beta\, N_H^+)\, v_H^- \;=\; 0, \tag{11.2}$$

$$\rho_{0,t} + (\rho_0\, v_H^+) \;=\; 0, \tag{11.3}$$

$$N_{H,t}^+ + (v_H^+\, N_H^+)_x - \frac{1}{m_H}(\alpha\, N_H^- - \beta\, N_H^+) \;=\; 0, \tag{11.4}$$

$$N_{H,t}^- + (v_H^-\, N_H^-)_x + \frac{1}{m_H}(\alpha\, N_H^- - \beta\, N_H^+) \;=\; 0, \tag{11.5}$$

where k is Boltzmann's constant, T is the absolute temperature of the moving medium, the stress σ is defined in [3].

It was shown in [7] that the solution of the basic equations reduces to finding the solution to the concentration N_H^+,

$$N_H^+ = N_0 + \tilde{N}_H, \tag{11.6}$$

where $\tilde{N}_H$ is the solution to the equation

$$\tilde{N}_{H,tt} - \; a_1\, \tilde{N}_{H,xx} + a_2 \tilde{N}_{H,t} - a_3 \tilde{N}_{H,xxt} +$$

$$a_4(\tilde{N}_H^2)_{xx} + a_5(\tilde{N}_H \tilde{N}_{H,xt})_x + a_6\, (\tilde{N}_{H,x} \tilde{N}_{H,t})_x = 0, \tag{11.7}$$

where

$$a_0 = \frac{m_H}{\alpha},\; a_1 = \frac{3kT\beta((F_0 - \gamma N_0)\, m_H + 2\alpha)}{2(F_0\, m_H + \alpha)^2 m_H}, a_2 = \frac{\alpha + \beta}{m_H},$$

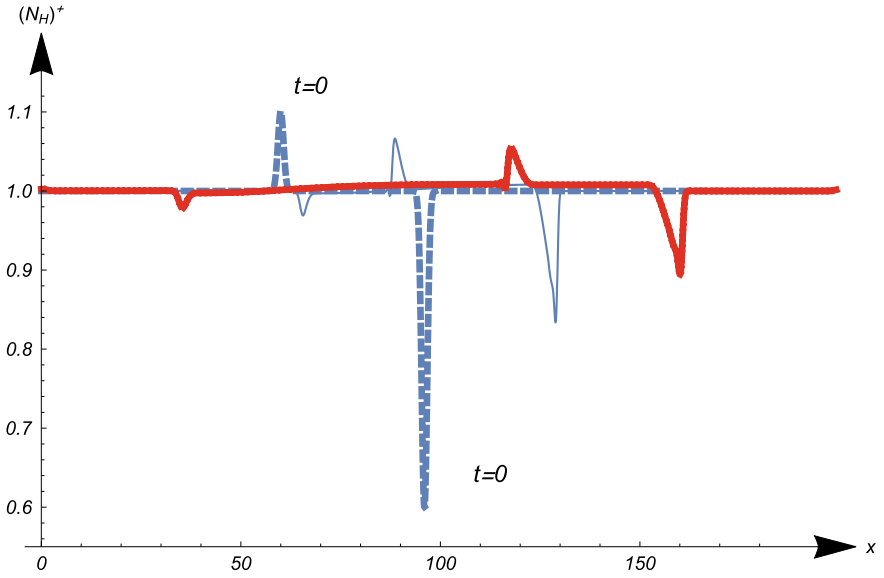

Fig. 11.1 Dynamics of concentration at the coefficients of Eq. (11.7): $a_1 = 1, a_2 = 0.01, a_3 = 0.01, a_4 = 0.75, a_5 = 0.075, a_6 = 0$. The profiles are shown in times 0, 30, 60

$$a_3 = \frac{3kTm_H((F_0 - \gamma N_0)\, m_H + 2\alpha - \beta)}{2(F\, m_H + \alpha)^2 m_H}, a_4 = \frac{3kT\gamma\beta}{4(F_0\, m_H + \alpha)^2}$$

$$a_5 = \frac{3kTm_H\gamma}{2(F_0\, m_H + \alpha)^2},\ a_6 = \frac{3kT(\alpha - \beta)}{2(F\, m_H + \alpha)^2 N_0}.$$

The physical meaning of the coefficients in Eq. (11.7) relates to an influence of the corresponding terms in the equation on the dynamics of concentration. Thus, the dominance of coefficient a_1 results in a hyperbolic wave nature of the propagation of the concentration front described by the terms with the second-order spatial and temporal derivatives, while the terms with coefficients a_2 and a_3 are responsible for the decay of the concentration wave. The coefficients $a_4 - a_6$ at nonlinear terms in Eq. (11.7) characterize the influence of nonlinearity, see [7] for details.

Numerical simulations demonstrate the influence of different nonlinear terms on the dynamics of localized variation in concentration. Numerical simulations are performed by assuming $N_0 = 1$ and considering evolution of a localized input defined by a Gaussian distribution,

$$\tilde{N}_H = N_0 + Q_1 \exp(-(x - x_0)^2/Q_2) + Q_3 \exp(-(x - x_1)^2/Q_2) \text{ at } t = 0, \tag{11.8}$$

where x_i and Q_i, are constants. A unidirectional evolution is provided by an extra nonzero initial condition for the initial velocity,

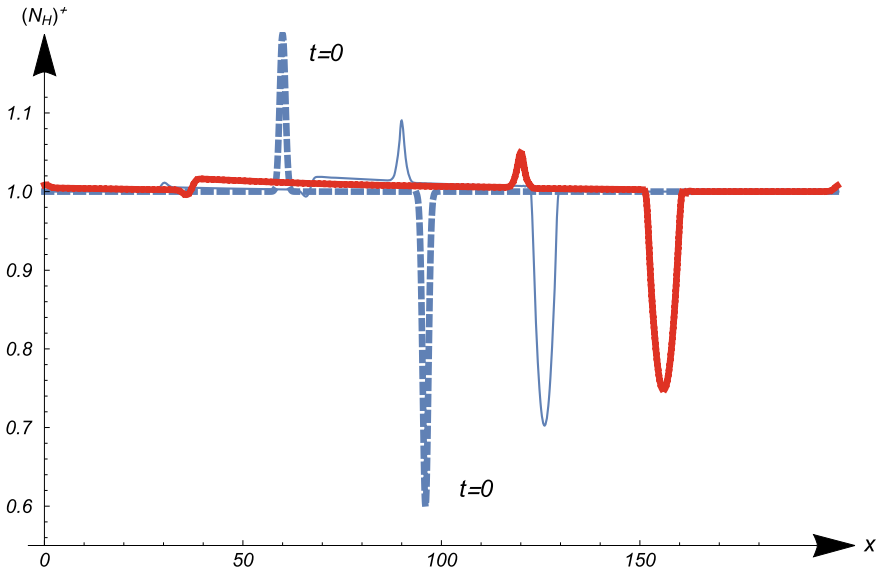

Fig. 11.2 Dynamics of concentration at the coefficients of Eq. (11.7): $a_1 = 1, a_2 = 0.01, a_3 = 0.01, a_4 = 0, a_5 = 0, a_6 = 0.5$. The profiles are shown in times 0, 30, 60

Shown in Fig. 11.1 is the evolution of the initial condition containing both negative and positive parts, Q_1 and Q_3 are of either sign in Eq. (11.8). They are marked by $t = 0$. One can see those initial disturbances propagate losing their amplitudes. Besides, a decrease in the amplitude, one can see a development of an asymmetry of the profile: the negative part suffers a steepness of the front edge of the wave, while the positive part demonstrates a steepness of the back edge of the wave.

On the contrary, an influence of the nonlinear term $(\tilde{N}_{H,x}\tilde{N}_{H,t})_x$ on the same initial condition doesn't provide an asymmetry as shown in Fig. 11.2. There is a decrease in the amplitude for both the positive and negative parts of the input, also one can see an increase in the width of the propagating disturbance of concentration.

More simulations about relative influence of nonlinearities and an influence of the polarity of the input on the behavior of the localized wave of concentration can may be found in Ref. [7].

11.3 Dicrete Continuum Nonlinear Model

The decrease in the amplitude of concentration wave happens due to an influence of dissipative terms in Eq. (11.7). The dynamical behavior of concentration varies when the strains in the material are taken into account.

The crystalline structure of a material can be described by a one-dimensional chain with masses connected by elastic springs and only with neighboring interactions between masses when the interaction obeys the Hookean linear law. Then the equation of motion for a mass with the number n is

$$m\ddot{u}_m = C_0(u_{n+1} - u_n) - C_0(u_n + u_{n-1}), \tag{11.9}$$

where u_i is the displacement of the i-th mass in the chain, m is the mass of the element of the chain, C_0 is the constant stiffness.

We assume a rheological relationship, $C = C_n(n, t)$,

$$\frac{N_0 + N_n}{C_n} = \frac{N_0}{C_0} + \frac{N_n}{C_H}, \tag{11.10}$$

where N_0 is the known constant concentration of the elements of the chain in the material, $N_n(n, t)$ is the discrete concentration of hydrogen, C_H is some known constant, $N_n(n, t)$ is the concentration of hydrogen in nanovoids, and C_H characterizes the weakening of the material due to the formation of nanovoids. Then we obtain

$$C_n = \frac{(N_0 + N_n)C_H C_0}{C_H N_0 + C_0 N_n}. \tag{11.11}$$

The statement of the problem is described more precisely in [8].

The continuum concentration $N(x, t)$ is usually described by an equation of transfer,

$$N_t + (\beta_0 + \beta_1 u_x)N + \tilde{\delta} u_x + \gamma N_{xx} = 0. \tag{11.12}$$

where a diffusion of the hydrogen concentration, N_{xx} is taken into account as well as a contribution of strains in the dynamics of concentration. First, it affects the source term coefficient $\beta_0 + \beta_1 u_x$, second, there is an influence δu_x to the variation of concentration. β_0, β_1, $\tilde{\delta}$, γ are constants.

The weakly long-wavelength case is considered and a small parameter ε is introduced,

$$u = \varepsilon u(X, T, , \tau, Z),\ y = \varepsilon^2 y(X, T, \tau, Z),$$

where

$$X = \varepsilon x,\ T = \varepsilon t,\ \tau = \varepsilon^3 t, ,\ Z = \varepsilon^4 t.$$

where

$$y(x, t) = \frac{C_0}{C_H}\frac{N}{N_0}.$$

It is shown in Ref. [8] that for small y one assumes

$$y = y_0 + \varepsilon y_1 + \ldots$$

and the governing continuum equation for y_0 is

$$2\,h\beta_0\sqrt{C_0 m}\,y_{0,\theta\tau} + \frac{\beta_0 C_0 h^4}{12}\,y_{0,\theta\theta\theta\theta} - \frac{\beta_0 C_0 h^2}{2}(y_0^2)_{\theta\theta} = 0. \tag{11.13}$$

where

$$\theta = X - \sqrt{\frac{C_0}{m}}\,T. \tag{11.14}$$

The traveling wave solution is [8]

$$y_0 = F - h^2\kappa^2 \mathrm{sech}^2(\kappa\xi), \tag{11.15}$$

where

$$\xi_\theta = 1, \xi_\tau = -W,\ W = \frac{h\sqrt{C_0}}{6\sqrt{m}}\left(h^2\kappa^2 - 3F\right)$$

The parameter W should be

$$W < -\frac{\kappa^2\, h\sqrt{m}}{3\sqrt{C_0}}.$$

to achieve only positive values of the solution for concentration.

Numerical results shown in Fig. 11.3 are the dynamics of an initial disturbance of concentration around constant value F. The initial profile contains both positive

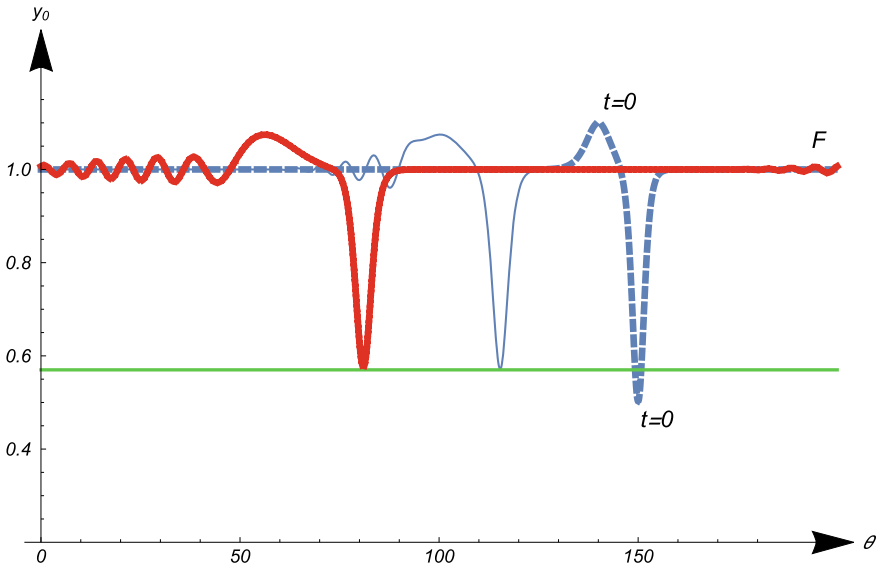

Fig. 11.3 Dynamics of concentration The constant $F = 1$

and negative amplitude disturbances marked by $t = 0$. As times goes on, the negative amplitude part evolves into traveling wave with a constant amplitude which is illustrated by a horizontal line in Fig. 11.3. The wave travels to the left. The positive amplitude disturbance is dispersed and doesn't give rise to a traveling localized wave of concentration.

The results obtained are written in the transformed coordinates. Coming back to the original variables, one obtains the solution for the stiffness [8]

$$C = C_0(1 - \varepsilon^2 F + \varepsilon^2\kappa^2 h^2 \mathrm{sech}^2(\varepsilon\kappa(x - (\sqrt{\frac{C_0}{m}} + \varepsilon^2 W)t). \tag{11.16}$$

The initial constant concentration F results in a decrease in the stiffness. The propagating wave of concentration locally increases the stiffness but not above C_0. The wave moves in the same direction as those shown in Figs. 11.1 and 11.2. More numerical solutions can be found in Ref. [8].

11.4 Conclusions

Several related phenomena, the formation of nanovoids associated with the hydrogen accumulation, the weakening of the material as a result of the nanovoids formation, and the non-uniform distribution of hydrogen and nanovoids, which is observed during the tension of corset specimens without stress concentrators with a uniform initial hydrogen concentration at a uniform uniaxial deformation, are described using the nonlinear wave propagation.

The dynamics or the relatively high rate of these processes can be explained by the fact that hydrogen itself does not move inside the material, but changes its state passing from a diffuse phase to a gaseous one inside nanovoids which can appear and disappear when the stress and strain change. Most likely, the consequences of this transformation for the strength and internal microstructure of the metal depend on the parameters of the material, loading rate, temperature, and value of the initial hydrogen concentration. The proposed wave approach makes it possible to establish these relations after identifying the parameters of the models.

Two models are discussed. The first one concerns the modeling of a bi-continuum. Numerical simulations reveal different influences of the nonlinear terms appearing in the presence or in absence of the inhomogeneous force. Qualitatively different effects such as arising of the tail behind the localized wave and formation of the counterpart wave are found. Also, the nonlinear discrete-continuum model of mutual influence of the hydrogen concentration and longitudinal strain in a chain is developed. A nonlinearity is introduced via the variations in the coefficient of stiffness of the springs between the elements of the chain. A two-dimensional lattice model will be the subject of our future work to describe both an increase and a decrease in the elastic constants.

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