

# The conditions of parametric instability in materials containing the hydrogen

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## Abstract

The full description of the hydrogen concentration change in volume and binding energy were done by two-continuum model. The main feature of this model is use of two energy state of the hydrogen. They are state with low binding energy and state with high binding energy. In state with high binding energy the hydrogen is moved with lattice of material. The hydrogen can freely move in the state with low binding energy. In this stage hydrogen may be modeled as a perfect fluid inside material.

The entire article is based on the experimental research work that study of the influence of the low hydrogen concentration upon mechanical properties of materials. It carried out analytical simulation of periodic uniaxial strain with taking into account of hydrogen with different binding energy. In the case of uniaxial deformation of material the equation of number hydrogen particle can be transformed in to Mathieu equation. This equation has instability area.

The investigation of this instability area had made. We determine the critical value of the hydrogen concentration and amplitude of the deformations. The model also gives the correlation among the hydrogen concentration, amplitude and frequency of the cyclic loading.

This result allows us to describe the process of destruction as instability caused by parametric resonance. Also, solution of this equation gives critical hydrogen concentration and safety level of loadings.

## 1 Introduction

The hydrogen is always present in metals and it has strong influence on mechanical properties. It accumulates in traps of various nature with various bond energy. The metals can get different properties depending on the hydrogen concentration and its bond energy.

The cyclic loading of material leads to redistribution of hydrogen in volume (Gorsky effect) [1] and redistribution of hydrogen in energy levels [2]. It can lead to the loss of strength. There are multiple cases of accidents, and situations when decreasing of the material resource related to hydrogen embrittlement.

## 2 Experiment

The experiment was carried out with samples of cylindrical form after cycles loading. The material of samples is aluminum alloy. The samples have cylindrical form with a truncation at the center. The samples were exposed by cycling loading in the testing machine with different loading and different number of cycles. Some of the samples were destroyed during test.

The hydrogen concentration was measured after mechanical testing in the different parts of samples. The samples were cut on the 5 parts for that. The hydrogen concentration was measured in the each part. Parameters of loading, number of cycles, state of the samples after loading and hydrogen concentration in the parts 1-4 are in the table 1. Here  $Q_1 - Q_4$  are hydrogen concentration in the different part of sample.  $Q_1$  – is hydrogen concentration in the area with minimum of section,  $Q_2 - Q_3$  – are the hydrogen concentrations in the area near from area 1,  $Q_4$  – is the hydrogen concentration in the non-deformed area.



Figure 1: The plan of the samples cutting

Table 1. Parameters of loading

Sample Num	Amplitude	Number of cycles	State of sample	$Q_1$	$Q_2$	$Q_3$	$Q_4$
	[MPa]	[MIO]		[ppm]	[ppm]	[ppm]	[ppm]
1	140	10	safe	0,49	0,47	0,46	0,6
2	160	5,44	safe	0,5	0,52	0,44	0,57
3	180	3,21	broken	0,94	0,73	0,44	0,62
4	180	0,84	broken	0,93	0,79	0,82	0,8
5	180	0,35	safe	0,74	1,04	0,84	0,81
6	0	0	original	0,4	0,6	0,58	0,57

Determination of the hydrogen carried out on the hydrogen analyzer AV-1. It works on the principle of vacuum-heating. The sample under investigation is heated in the vacuum under constant temperature. Extracted gases are analyzed using mass-spectra.

Figure 2 shows the distribution of hydrogen in the volume of samples. Curve 2 shows distribution of hydrogen for sample 3 (3 mln cycles, broken), and curve 1 shows distribution of hydrogen for sample 6 (original, 0 cycles)

It is seen that distribution of hydrogen in the curve 1 is evenly but for point 1. Low concentration of hydrogen in the point 1 can be accounted for the sample preparation for the analyses of hydrogen definition. Compare curve 2 and curve 1. It is seen that, hydrogen concentration in the area 4 is equally, but in the points 3 the hydrogen concentration in the curve 2 lower than curve 1, and in the point 2 and 1 it is higher. It looks like transfer of the hydrogen from area 3 to area 2 and 1. There is some process redistribution of hydrogen during cycles loading. The result this process is decreases hydrogen concentration in the same area and increases hydrogen concentration in the neighboring area. The similar process is found in the all samples after cycles loading. There is some correlation between amount of hydrogen concentration and number of cycles.

### 3 Modeling

The full description of the change hydrogen concentration in volume and binding energy were done in two continuum model [3]. The main feature of this model is use of two energy state of the hydrogen. They are state with low binding energy and state with high binding energy. In state with high binding energy the hydrogen is moved with lattice of

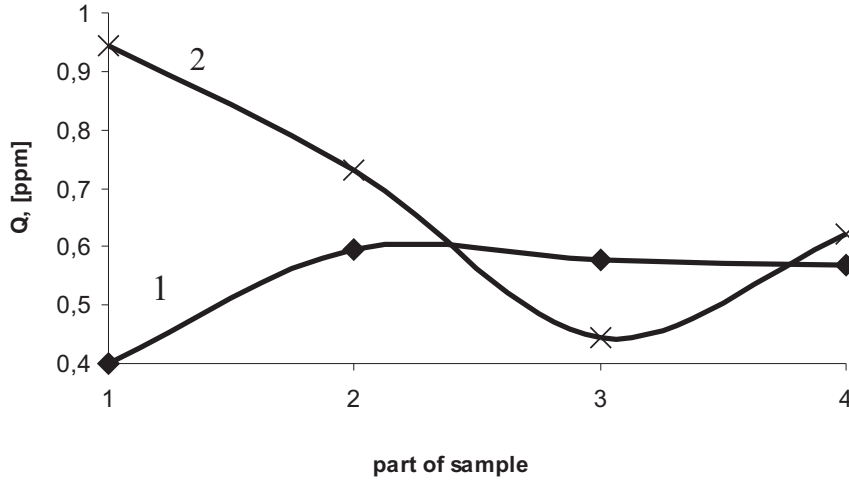


Figure 2: Distribution of hydrogen in the volume of samples

material. In the state with low binding energy the hydrogen can freely move in volume. It is modeled as a perfect fluid inside material.

The equation of motion of the lattice:

$$\begin{aligned}\frac{\partial \sigma^{(1)}}{\partial x} &= \rho^{(1)} \frac{\partial v^{(1)}}{\partial t} + J_{12} v^{(1)} + R_{12}, \\ \rho^{(1)} &= \rho^{(0)} + \rho_H^{(+)}, \\ \rho^{(0)} &\gg \rho_H^{(+)}\end{aligned}$$

where  $\rho_H^{(+)}$  is the density of attached particles of hydrogen,  $\rho^0$  is the density of material,  $J_{12}v^{(1)}$  is the reaction force, this force relate to attached mobile hydrogen with lattice,  $R_{12}$  is the internal force of interaction between lattice and mobile hydrogen

The equation of motion of mobile hydrogen is:

$$\begin{aligned}-\frac{\partial p}{\partial x} &= \rho^{(2)} \frac{\partial v^{(2)}}{\partial t} + J_{12} v^{(2)} + R_{21}, \\ \rho^{(2)} &= \rho_H^{(-)} = m_H \cdot N^{-}\end{aligned}$$

where  $N^{-}$  is the number of particle of mobile hydrogen.

The relation between pressure and density of mobile hydrogen is:

$$p - p_0 \cong \rho_H^{(-)} \cdot C_H^2 = m_H N^{-} \cdot C_H^2.$$

Mass balance equation attached hydrogen particles is:

$$\frac{\partial N_H^+}{\partial t} + \frac{\partial (N_H^+ v^{(1)})}{\partial x} = \frac{J_{12}}{m_H}.$$

Mass balance equation free hydrogen particles is:

$$\frac{\partial N^-}{\partial t} + \frac{\partial (N^- v^{(1)})}{\partial x} = \frac{J_{21}}{m_H}.$$

Where  $J_{12} = -J_{21} = \alpha N^- - \beta N_H^+$ ,  $\alpha, \beta$ - is some constant.

The mobile hydrogen is moved inside material like perfect liquid, than:

$$R_{12} = -R_{21} = k \frac{N^- m_H}{D(\varepsilon)} \left( v^{(2)} - v^{(1)} \right).$$

where  $D(\varepsilon)$  is the size of channel of diffusion, it depends on deformation  $\varepsilon$ , and  $k$  is the linearization coefficient.

In the case of uniaxial deformation of material the equation of bound hydrogen particles balance is:

$$\frac{\partial^2 n_H^+}{\partial t^2} + (\alpha + \beta) \frac{\partial n_H^+}{\partial t} - \frac{C_H^2}{k} D(\varepsilon_{st}) \left[ \beta \frac{\partial^2 n_H^+}{\partial x^2} + \frac{\partial^3 n_H^+}{\partial t \partial x^2} \right] = 0. \quad (1)$$

Use the Fourier method:  $n_H^+(t, x) = T_{n_H^+}(t) \cdot X_{n_H^+}(x)$ .

Then

$$\frac{\ddot{T}_{n_H^+}(t) + (\alpha + \beta) \dot{T}_{n_H^+}(t)}{\frac{C_H^2}{k} D(\varepsilon_{st}) (\beta T_{n_H^+}(t) + \dot{T}_{n_H^+}(t))} = \frac{X_{n_H^+}''(x)}{X_{n_H^+}(x)}.$$

The equation for function  $X_{n_H^+}(x)$  is:

$$X_{n_H^+}''(x) + \gamma_x^2 X_{n_H^+}(x) = 0.$$

The equation for function  $T_{n_H^+}(t)$  is:

$$\ddot{T}_{n_H^+}(t) + (\alpha + \beta + \gamma_x^2 \frac{C_H^2}{k} D(\varepsilon_{st})) \dot{T}_{n_H^+}(t) + \gamma_x^2 \frac{C_H^2}{k} D(\varepsilon_{st}) \beta T_{n_H^+}(t) = 0. \quad (2)$$

Let deformation is  $\varepsilon = \varepsilon_1 \cdot \cos(\omega t)$  and  $D(\varepsilon) = D_0 + D_1 \cdot \varepsilon$  than:

$$\ddot{T}_{n_H^+}(t) + \left[ \alpha + \beta + (G_0 + G_1 \cos(\omega t)) \right] \dot{T}_{n_H^+}(t) + (G_0 + G_1 \cos(\omega t)) \beta T_{n_H^+}(t) = 0 \quad (3)$$

or

$$\ddot{T} + 2\Gamma(1 + \gamma + 2\mu \cos \omega t) \dot{T} + \Omega^2(1 + 2\mu \cos \omega t) T = 0. \quad (4)$$

Where  $2\mu = G_1/G_0$ ,  $2\Gamma = G_0$ ,  $\gamma = (\alpha + \beta)/G_0$ .

The equation (4) looks like Mathieu equation. Mathieu equation has instability area. The solution will be found in the form:

$$T(t) = a \sin\left(\frac{\omega t}{2}\right) + b \cos\left(\frac{\omega t}{2}\right). \quad (5)$$

Substituting (5) into (4) we get

$$\begin{aligned} & -\frac{1}{4}a\omega^2 \cos\left(\frac{\omega t}{2}\right) - \frac{1}{4}b\omega^2 \sin\left(\frac{\omega t}{2}\right) - \\ & -\Gamma\omega a(1 + \gamma + 2\mu \cos(\omega t)) \sin\left(\frac{\omega t}{2}\right) + \Gamma\omega b(1 + \gamma + 2\mu \cos(\omega t)) \cos\left(\frac{\omega t}{2}\right) - \\ & + \Omega^2 a \cos\left(\frac{\omega t}{2}\right) + \Omega^2 b \sin\left(\frac{\omega t}{2}\right) + 2\Omega^2 \mu a \cos\left(\frac{\omega t}{2}\right) \cos(\omega t) + \\ & + 2\Omega^2 \mu b \cos\left(\frac{\omega t}{2}\right) \sin(\omega t) = 0. \end{aligned} \quad (6)$$

Conversion products of trigonometric functions into a sum leads to the equation

$$\begin{aligned} & \left( a \left[ -\omega\Gamma(1 + \gamma - \mu) \right] + b \left[ -\frac{\omega^2}{4} + \Omega^2(1 - \mu) \right] \right) \sin\left(\frac{\omega t}{2}\right) + \\ & + \left( a \left[ -\frac{\omega^2}{4} + \Omega^2(1 + \mu) \right] + b \left[ \Gamma\omega(1 + \gamma + \mu) \right] \right) \cos\left(\frac{\omega t}{2}\right) = 0. \end{aligned} \quad (7)$$

Since both  $\sin \omega t/2$  and  $\cos \omega t/2$  are linearly independent functions, both their coefficients in equation (7) must vanish. This gives a system of two linear equations in a and b

$$\begin{cases} a [-\omega\Gamma(1 + \gamma - \mu)] + b \left[-\frac{\omega^2}{4} + \Omega^2(1 - \mu)\right] = 0 \\ a \left[-\frac{\omega^2}{4} + \Omega^2(1 + \mu)\right] + b [\Gamma\omega(1 + \gamma + \mu)] = 0 \end{cases} \quad (8)$$

Nontrivial solution of (8) are possible only if the determinant of the system is zero, that is,

$$\det = \Gamma^2\omega^2 \left( (1 + \gamma)^2 - \mu^2 \right) + \left( -\left(\frac{\omega}{2}\right)^2 + \Omega^2 \right)^2 - \mu^2\Omega^4 = 0. \quad (9)$$

From equation (9) can be obtained the functional dependence  $\mu = \mu(\omega)$

$$\mu^2 = \frac{\left[ \Omega^2 - \left(\frac{\omega}{2}\right)^2 \right]^2 + [\Gamma\omega(1 + \gamma)]^2}{\Omega^4 + \Gamma^2\omega^2} \quad (10)$$

where  $\Omega = \sqrt{G_0\beta} = C_H\gamma_x\sqrt{\frac{D_0\beta}{k}}$  is the “fundamental frequency” of the hydrogen concentration in the material. Upon the application of time of fatigue, we can achieve by hydrogen accumulation ratio  $\omega = \frac{\Omega}{2}$ . This frequency is critical. At this frequency occurs parametric resonance.

Analysis of equation (10) shows that the stability boundary in the plane  $(\mu, \omega)$  has a minimum in the neighborhood of  $\omega = 2\Omega$  subjected to

$$2\frac{\Omega^2}{\beta^2} + 1 > \frac{\alpha + \beta}{\beta} \left( 2\frac{\Omega^2}{\beta^2} + \frac{\alpha + \beta}{\beta} \right).$$

This inequality is satisfied only if  $(\alpha + \beta)/\beta < 1$ . This ratio the coefficients of linearization is typical for the alloys accumulating diffusely mobile hydrogen. Part of the dependence  $\mu = \mu(\xi)$  for  $\xi \geq 0$ , where  $\xi = \frac{\omega}{\Omega}$ , is shown in figure 3.

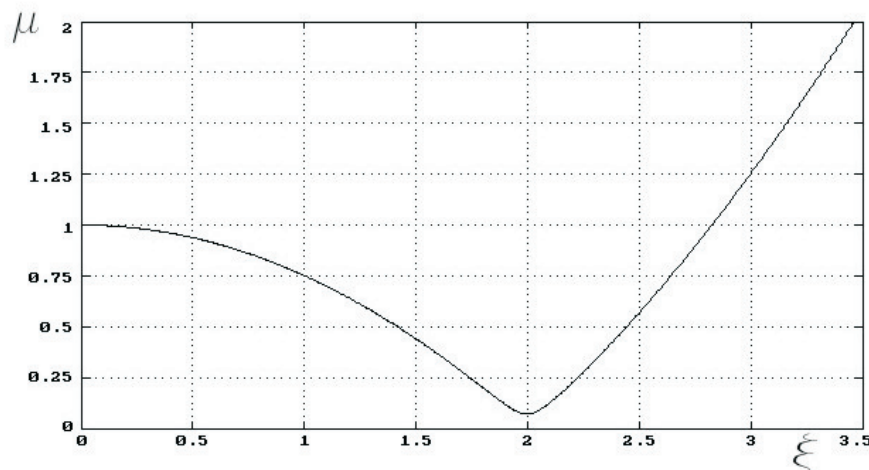


Figure 3: Instability area of equation

This result allows us to describe the process of destruction as instability caused by parametric resonance. Also, solution equation 1 gives critical hydrogen concentration and safety level of loadings, when there is not destruction.

## 4 Conclusions

It is shown, that during periodical loading is some process. This process leads to redistribution of hydrogen in volume of material. The accumulation of the hydrogen occurs in the area with maximum of tension. Besides, accumulation of hydrogen in the one area is as result of decrease hydrogen concentration in neighbor area. This fact allows to make a conclusion that hydrogen at first redistributed within the material, and then starts to diffuse from the outside.

On the basis of two-continuum model carried out simulation of process uniaxial cycling loading. During simulation was obtained the balance of hydrogen equation. The stability domain is obtained during solution of the equation. The stability domain allows to define critical frequency and safe level of loading.

## Acknowledgements

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