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**RESEARCH REPORT**  
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# Simulation by a diffusion model for the variation of hydrogen pressure with time between hydrogen discharge shots in LHD

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## Abstract

When hydrogen gas discharge is carried out in a magnetic fusion experimental device, hydrogen is implanted into the first wall during discharge but is in turn released from the wall in the pause period of discharge shot. To express the hydrogen desorption rate from the wall as a function of time, a diffusion model is presented, in which the transport of trapped hydrogen in the wall is treated by the one dimensional diffusion equation. It is discussed that when the deposition profile of implanted hydrogen in the wall is given as a function of position, the diffusion equation can be solved analytically. The hydrogen desorption rate is numerically calculated using the analytical solution, then it is shown that the calculated desorption rate well explains the experimental hydrogen desorption rate which has been observed as a function of time after hydrogen discharge shot in LHD.

Keywords: H trapping and desorption, diffusion, recombination, stainless steel wall, LHD

## 1. Introduction

For the confinement study of plasma in LHD hydrogen pulse discharge for the duration of 3 or 5 s is carried out every 10 min. However, when a high density hydrogen plasma of the order of  $10^{20} \text{ m}^{-3}$  is produced, we have occasionally to wait for a time longer than 10 min to shot the next pulse discharge, because hydrogen desorption lasts with a very high rate from the first wall of the plasma vacuum vessel. Therefore, it is an urgent problem to consider how to shorten the time interval for the repetition of pulse discharge. So in this paper a diffusion model is proposed to explain the desorption mechanism of hydrogen from the wall surface after hydrogen pulse discharge. In the proposed model, it is assumed that energetic hydrogen atoms generated at plasma boundary during hydrogen pulse discharge in LHD are implanted into the first wall, in which implanted hydrogen atoms are once trapped in the interstitial sites of the solid wall but soon are detrapped because of low trapping energy and become mobile

hydrogen atoms, as a result, in the pausing period of discharge the mobile hydrogen atoms which arrive to the wall surface due to diffusion are desorbed to the gas phase as hydrogen molecules through recombination reaction. To calculate theoretically the desorption rate of hydrogen, the transport of hydrogen atoms in the solid wall is considered using the one dimensional diffusion equation. If we can know the density distribution function of trapped atoms in the solid wall at time  $t=0$  right after pulse discharge as a function of position, we may construct the boundary and initial conditions to solve the diffusion equation. As a trial, by referring the density distribution functions which have been calculated by Bastasz et al.,[1,2] using Trim code for the cases when low energy H ions from 100 to 450 eV are implanted into Pd target, we may assume a density distribution function of hydrogen implanted in the first wall of LHD. As a result, using the solution of the diffusion equation it is shown that the hydrogen desorption rate after discharge shot in LHD can be expressed as a function of time.

In this paper first the construction of the diffusion model is described. Secondly the solution of diffusion equation is derived. Thirdly the results of numerical calculation for hydrogen density, hydrogen diffusion flux, and density distribution function are introduced. Finally the comparison between calculation and experiment for hydrogen desorption rate is discussed.

## 2. Diffusion model for hydrogen desorption

When hydrogen gas discharge is carried out in a plasma device, energetic hydrogen atoms generated at the boundary region of plasma are implanted into the first wall of the plasma device, where a part of incident hydrogen atoms is reflected from the wall, but the other part is trapped in the wall. When hydrogen ions with energies of 100, 250, and 400 eV are implanted into a palladium target, the distribution ranges of trapped hydrogen in the target have been calculated using Trim code by Bastasz[1,2], and the calculated results show that trapped hydrogen atoms distribute in the depth within 30 nm and the hydrogen density distribution function  $f(x)$  as a function of trapped position  $x$  of hydrogen in the target has a maximum between two positions of  $x=0$  and  $x=a$ , where  $a$  is the maximum depth of trapped hydrogen atom in the target. Therefore, if the density distribution

function takes a maximum at  $x = R$  (where  $0 < R < a$ ), we may consider that the maximum value  $f(R)$  will gradually increase with the repetition of the hydrogen pulse discharge and finally reaches a saturation level, because the trapped hydrogen atoms are mobile in the bulk and diffuse out to the surface and the depth of solid during pause of discharge with time.

Now to predict the hydrogen desorption rate after discharge shot by a diffusion model, we need to consider first the density change of hydrogen with time at the wall surface of  $x=0$ . By letting the hydrogen surface density as  $u_s(t) (\text{cm}^{-3})$ , we may express the conservation of mass equation for hydrogen atoms at the wall surface as follows;

$$\delta \frac{du_s}{dt} = -2Ku_s^2 + s(1-\theta)\Gamma, \quad (1)$$

where the first and second terms in the right-hand side represent the hydrogen desorption rate as molecules due to recombination reaction at the surface and the adsorption rate of hydrogen atoms which arrive to the surface due to diffusion,  $\delta(\text{cm})$  is the distance between neighboring interstitial sites wherein dissolved hydrogen atoms can be located,  $K(\text{cm}^4\text{s}^{-1})$  is the recombination coefficient of adsorbed atoms,  $s$  is the sticking probability of a hydrogen atom to vacant adsorption sites,  $\theta$  is the fractional coverage defined with the maximum adsorption density  $u_m(\text{cm}^{-3})$  as

$$\theta = \frac{u_s}{u_m}, \quad (2)$$

and  $\Gamma$  is the diffusion flux of hydrogen atoms from the bulk to the surface which is expressed with the diffusion coefficient  $D(\text{cm}^2\text{s}^{-1})$  and the hydrogen density  $u(x,t)$  in the bulk as follows;

$$\Gamma = D \left\{ \frac{\partial u(x,t)}{\partial x} \right\}_{x=0}. \quad (3)$$

## 2.1 Density change in the transient phase right after discharge

In the transient phase right after discharge shot, we can expect that the adsorption sites of the wall surface are fully occupied by the implanted hydrogen atoms. This means that the coverage  $\theta$  increases to a high value near 1 and so the adsorption probability  $s(1-\theta)$  becomes very small. For such situation we may solve eq. (1) by assuming that the adsorption term of  $s(1-\theta)\Gamma$  is constant. Then by letting the initial surface density as  $u_s(0) = u_0$  at  $t=0$ , we obtain the following solution;

$$u_s(t) = \rho \left( \frac{1 + He^{-\frac{4K\rho}{\delta}t}}{1 - He^{-\frac{4K\rho}{\delta}t}} \right), \quad (4a)$$

where

$$\rho = \sqrt{\frac{\varepsilon\Gamma}{2K}} \quad (4b)$$

for  $0 < \varepsilon = s(1 - \theta) \ll 1$  and  $\varepsilon\Gamma = \text{constant}$ , and

$$H = \frac{u_0 - \rho}{u_0 + \rho}. \quad (4c)$$

Eq. (4a) shows that in the transient phase right after discharge shot, the surface density decreases or increases exponentially with the time constant of  $\delta/4K\rho$  according to that  $u_0 > \rho$  or  $u_0 < \rho$  and reaches the constant value  $\rho$  for  $t > \delta/4K\rho$ .

## 2.2 Density change in a quasisteady state

In the quasisteady state in which the condition  $du_s/dt \cong 0$  in eq. (1) holds, because vacant sites at the wall surface starts to increase with the decrease of surface density, then the hydrogen atoms which arrive at the surface due to diffusion start to adsorb on the vacant sites. Thus we may express the hydrogen desorption rate from the wall surface as follows;

$$(2Ku^2(x,t))_{x=0} = s(1 - \theta(t)) \left( D \frac{\partial u(x,t)}{\partial x} \right)_{x=0}, \quad (5a)$$

where using eq. (2)  $\theta(t)$  is given by

$$\theta(t) = \frac{u(0,t)}{u_m}. \quad (5b)$$

Eq. (5a) represents the first boundary condition at  $x = 0$  for the following one dimensional diffusion equation;

$$\frac{\partial u}{\partial t} = D \frac{\partial^2 u}{\partial x^2}. \quad (6)$$

Using the solution  $u(x,t)$  of eq. (6) which describes the density of mobile hydrogen atoms as functions of time  $t$  and position  $x$  in the bulk of the wall, we can predict the hydrogen desorption rate at the wall surface  $x = 0$  as  $Ku^2(0,t)$ . To solve eq. (6), we need more the initial condition at time  $t = 0$  and the second boundary condition at  $x = a$ . Here let us assume those conditions as follows;

$$u(x,t) = f(x) \quad \text{at } t = 0, \quad (7)$$

and

$$u(x,t) = 0 \quad \text{at } x = a. \quad (8)$$

In eq. (7), we assume that the distribution function  $f(x)$  can be expressed as follows;

$$f(x) = \alpha(1 + \beta x) + \gamma \left( e^{-\frac{x}{b_1}} - e^{-\frac{x}{b_2}} \right), \quad (9)$$

where  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $b_1$ , and  $b_2$  are constants, and we may determine values of these constants if we can calculate the range distribution function of hydrogen implanted in a stainless steel wall by using a simulation code such as Trim code. The boundary condition of eq. (8) means that the diffused atoms is completely diluted in the region of  $a < x$ , namely the reflection of diffused atoms to the backward region of  $x < a$  does not exist.

### 2.3 Solution of diffusion equation

It is well known that the top surface of a stainless steel used as a structural material for vacuum chamber is covered with oxide layer. Redhead [3] has suggested that under the presence of the oxide layer the adsorbed amount of hydrogen on the stainless steel surface is suppressed very low by the co-adsorption of other gases such as water vapor and carbon monoxide which usually exist as residual gases in a vacuum system. Therefore, we may estimate that the surface density of hydrogen atoms at the wall surface of a pumped vacuum chamber is around  $10^{20} \text{ cm}^{-3}$ . Thus we may set the maximum adsorption density for hydrogen as  $u_m \equiv 10^{20} \text{ cm}^{-3}$ . In addition, let us assume that the sticking probability  $s$  can be treated as  $s=1$  and the surface coverage  $\theta$  changes with time in the region of around  $\theta=1/2$  under the condition of quasisteady state. Then because we can treat the adsorption probability as

$$s(1-\theta) = \frac{u}{u_m}(1/\theta-1) \equiv \frac{u}{u_m} \quad \text{for } s=1 \quad \text{and} \quad \theta=1/2,$$

eq. (5a) can be simplified as follows:

$$\left\{ h \left( \frac{K}{D} \right) u - \frac{\partial u}{\partial x} \right\}_{x=0} = 0 \quad (10a)$$

where

$$h = 2u_m. \quad (10b)$$

By combining eqs, (7),(8),(9), and (10a), we obtain the following solution for the diffusion equation of eq. (6);

$$u(x,t) = \sum_{n=1}^{\infty} C_n e^{-D\lambda_n^2 t} \sin\{\lambda_n(x-a)\}, \quad (11a)$$

where  $\lambda_n$  is the eigen value determined from the following relation

$$\lambda_n = -\tan(\lambda_n a) \cdot \frac{ahK}{D} \quad (11b)$$

and  $C_n$  is the constant determined as

$$C_n = \int_0^a f(x) \sin\{\lambda_n(x-a)\} dx / \int_0^a \sin^2\{\lambda_n(x-a)\} dx. \quad (11c)$$

## 2.4 Numerical calculation

If it is allowable to use the calculated result of the distribution range of implanted H in Pd target shown by Bastasz[1,2], because the maximum values of density distribution functions for incident hydrogen ion energies from 100 to 450 eV appear at nearly the same position of  $x(=R)=5$  nm, we may set the values of constants  $b_1$ ,  $b_2$  in  $f(x)$  of eq. (9) as  $b_1=7$  and  $b_2=4$  nm, respectively. By referring the review on H trapping, diffusion and recombination by Langley [4], we may select the values of diffusion and recombination coefficients for hydrogen as  $D=7.7 \times 10^{-16} \text{cm}^2 \text{s}^{-1}$  and  $K=10^{-28} \text{cm}^4 \text{s}^{-1}$ . As a result, if we set the value of  $\alpha$  as  $\alpha=2$ , other two constants  $\beta$  and  $\gamma$  in  $f(x)$  can be determined as  $\beta=-0.14066$  and  $\gamma=487.474$  using eqs. (9) and (10a) for  $h=2u_m=2 \times 10^{20} \text{cm}^{-3}$ . Figure 1 shows the calculated density change of mobile hydrogen  $u(x,t)$  as a function of position when the elapsing time is changed every 360 s from 0 to 3600 s after discharge shot. The density change  $u(0,t)$  for elapsing time in Fig. 1 is shown in Table 1. Figure 2 shows the change of  $\partial u(x,t)/\partial x$  as a function of position when the elapsing time is changed every 360 s from 0 to 3600 s after discharge shot. Figure 3 shows the desorption rate of  $\text{H}_2$  molecules  $Ku^2(0,t)$  as a function of time after discharge shot.

## 3. Hydrogen pressure change after hydrogen discharge shot in LHD

In LHD the hydrogen desorption after discharge shot is observed as the hydrogen pressure change with time in the plasma vacuum vessel which is monitored by a quadrupole mass analyzer and an ionization gauge. The hydrogen desorption rate  $q(t)$  can be expressed with the measured hydrogen pressure as a function of time  $P_{\text{H}_2}(t)$  after discharge shot as follows;

$$q(t) = \frac{S}{A} \left[ \frac{P_{\text{H}_2}(t)}{kT} \right] (\text{molecules} \cdot \text{cm}^{-2} \text{s}^{-1}), \quad (12)$$

where  $S(\text{cm}^3 \text{s}^{-1})$  is the pumping speed for hydrogen in the pumping system of LHD,  $A(\text{cm}^2)$  is the wall surface area of the plasma vacuum vessel and  $kT$  is the conversion factor of the pressure to the gas density expressed as

$$(1/kT) = 3.24 \times 10^{16} \text{ molecules} \cdot \text{cm}^{-3} \text{Torr}^{-1} \text{ for } T = 298 \text{ K.}$$

We can consider that the experimental desorption rate of eq. (12) corresponds to the theoretical one as follows;

$$q(t) \Leftrightarrow Ku^2(0,t). \quad (13)$$

Because the ratio  $S/A$  in eq. (12) is constant, if we compare only the time dependence of desorption rates between theory and experiment, we can treat the experimental desorption rate as  $q(t) \propto P_{\text{H}_2}(t)$ . Figure 4 shows a typical pressure change  $P_{\text{H}_2}(t)$  measured as a function of time after discharge shot, which was quoted from the pumping down curve (data No.991124) in the third cycle period of plasma experiment in LHD.

#### 4. Discussion

From the comparison between Fig. 3 and Fig. 4, the decay constants of measured and calculated hydrogen desorption rates in the time region of  $0 < t < 600 \text{ s}$  are  $\tau(\text{measured}) = 250 \text{ s}$  and  $\tau(\text{calculated}) = 350 \text{ s}$ , respectively. This result shows that our diffusion model well explains the measured desorption rate. If we select carefully numerical values for diffusion and recombination coefficients so as to fit the calculated desorption rate to the measured one, we will be able to improve the accuracy of the numerical simulation. An interest is that the hydrogen diffusion coefficient adopted in this study is the surface layer diffusion coefficient, which is different from the bulk diffusion coefficient. Concerning with the magnitude of diffusion coefficient, it has been shown by Langley [4] that  $10^{-13} \leq D_{\text{bulk}} \leq 10^{-12} \text{ cm}^2\text{s}^{-1}$  for 304 or 316 stainless steel at room temperature and the surface layer diffusion coefficient becomes smaller than that by about  $2 \times 10^{-3}$  (i.e.,  $2 \times 10^{-16} \leq D_{\text{surface layer}} \leq 2 \times 10^{-15} \text{ cm}^2\text{s}^{-1}$ ). Therefore, we may say that the hydrogen desorption observed after discharge shot in LHD may be explained by the surface layer diffusion of hydrogen. As the surface layer on the first wall in LHD, we may consider a titanium gettered film which is made by flashing of Ti-ball for impurity control in LHD and may expect that the titanium gettered surface is naturally oxidized because of the impurity gas adsorption such as  $\text{H}_2\text{O}$  or  $\text{CO}$ .

Using the pumping speed  $S_{\text{H}_2} = 7 \times 10^7 \text{ cm}^3\text{s}^{-1}$  which is allotted for hydrogen in the pumping system of LHD and the surface area of the plasma vacuum vessel  $A = 5 \times 10^6 \text{ cm}^2$ , we can estimate the magnitude of hydrogen surface density at  $t = 0$  after discharge shot from eqs. (12) and (13) as



$$u(0,0) = \sqrt{\frac{S_{H_2} P_{H_2}(0)}{KAkT}} \cong 9.4 \times 10^{19} \text{ cm}^{-3}.$$

For the calculation of the hydrogen density by eq. (11) we used the distribution function  $f(x)$  expressed with dimensionless scale. Therefore, using eq. (7) if we assume that  $f(0) = \alpha$  ( $= 2 \text{ div}$ ) is equal to the above experimental density  $u(0,0) \cong 10^{20} \text{ cm}^{-3}$ , the density scale of the vertical axis in Fig. 1 may be given as  $5 \times 10^{19} \text{ cm}^{-3} / \text{div}$ . Then, from Fig. 1 the maximum density of trapped hydrogen  $f(R) = u(R,0)$  (where  $R = 5 \text{ nm}$ ) at  $t=0$  right after discharge shot may be estimated as

$$u(R,0) \cong 100 \text{ div} \times 5 \times 10^{19} \text{ cm}^{-3} / \text{div} = 5 \times 10^{21} \text{ cm}^{-3},$$

which value is 50 times larger than the surface density  $u(0,0)$  at  $x=0$ . In the pause period of discharge the maximum density  $u(R,t)$  starts to decrease with time due to desorption, and it decreases to around  $4 \times 10^{21} \text{ cm}^{-3}$  after 600 s ( $= 10 \text{ min}$ ). Thus as long as the pulse discharge is repeated every 10 min, we can say that almost all of trapped hydrogen still remains in the wall without releasing. In Fig. 2, we can see that the sign of  $\partial u / \partial x$  is reversed in the left- and right-hand sides of the position  $x=5 \text{ nm}$ . The plus sign of  $\partial u / \partial x$  shows that hydrogen diffuses backward, namely to the direction of surface. Therefore we can see that only the trapped hydrogen in the shallow region of  $0 < x < 5 \text{ nm}$  of the surface layer mainly contributes to desorption in the pause period of discharge.

In this study although the plasma vacuum vessel of LHD is fabricated from type 316 stainless steel, we refereed the calculated hydrogen depth profiles for Pd target with Trim code by Bastasz to simulate the hydrogen desorption rate in LHD. This is because we have no available data of hydrogen depth profile for a stainless steel target. So it is a future matter to calculate the hydrogen depth profile for a stainless steel target using any simulation code.

## 5. Conclusion

A diffusion model is proposed to explain the variation of hydrogen desorption rate with time which has been observed after pulse discharge shot in LHD. In the model the density change of implanted hydrogen into the wall is described by the one dimensional diffusion equation. As a result that the diffusion equation was solved under proper boundary and initial conditions, it was pointed out that the transport of implanted hydrogen in

conditions, it was pointed out that the transport of implanted hydrogen in the wall is not described by the bulk diffusion but rather described by the surface layer diffusion. It was also demonstrated that the calculated hydrogen desorption rate by the diffusion model well explains the observed hydrogen desorption rate in LHD. To simulate more accurately the hydrogen desorption rate in LHD, it is necessary to calculate the distribution range of hydrogen implanted in a stainless steel target with various ion energies using a simulation code.

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# Figure captions

- Figure 1.  $u(x,t)$  vs  $x$  when elapsing time is changed from 0 to 3600 s every interval of 360 s after discharge shot.
- Figure 2.  $\partial u(x,t)/\partial t$  vs  $x$  when elapsing time is changed from 0 to 3600 s every interval 360s after discharge shot.
- Figure 3.  $Ku^2(0,t)$  vs  $t$  after discharge shot.
- Figure 4. Hydrogen pressure change measured after discharge shot in LHD.
- Table 1. Change of surface density  $u(0,t)$  when elapsing time is changed from 0 to 3600 s in the curve of Fig. 1.

Table 1

time											
t(s)	0	360	720	1080	1440	1800	2160	2520	2880	3240	3600
density (div)											
u(0,t)	2.0	1.0	0.78	0.65	0.55	0.50	0.45	0.38	0.36	0.34	0.30

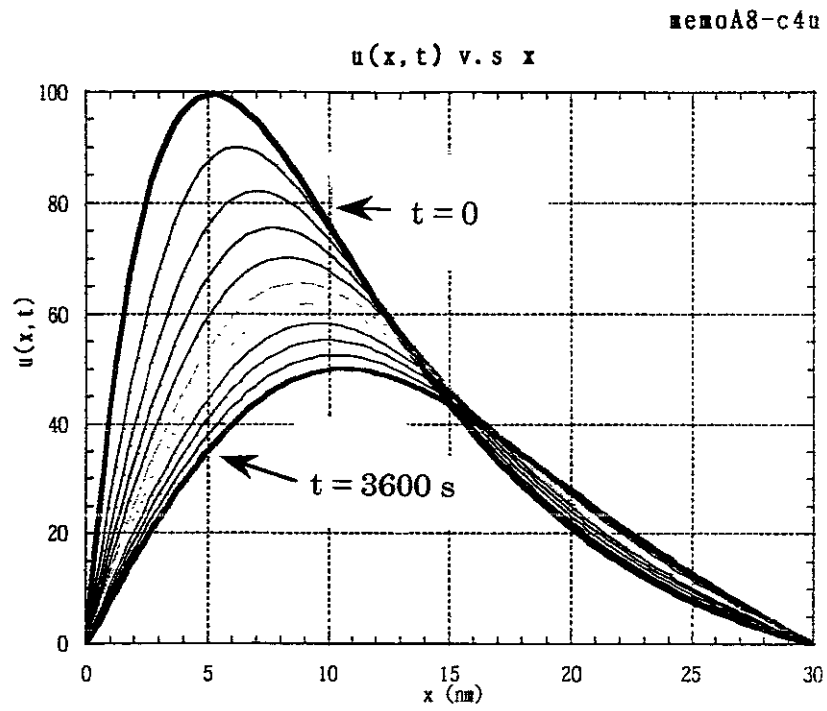


Fig. 1

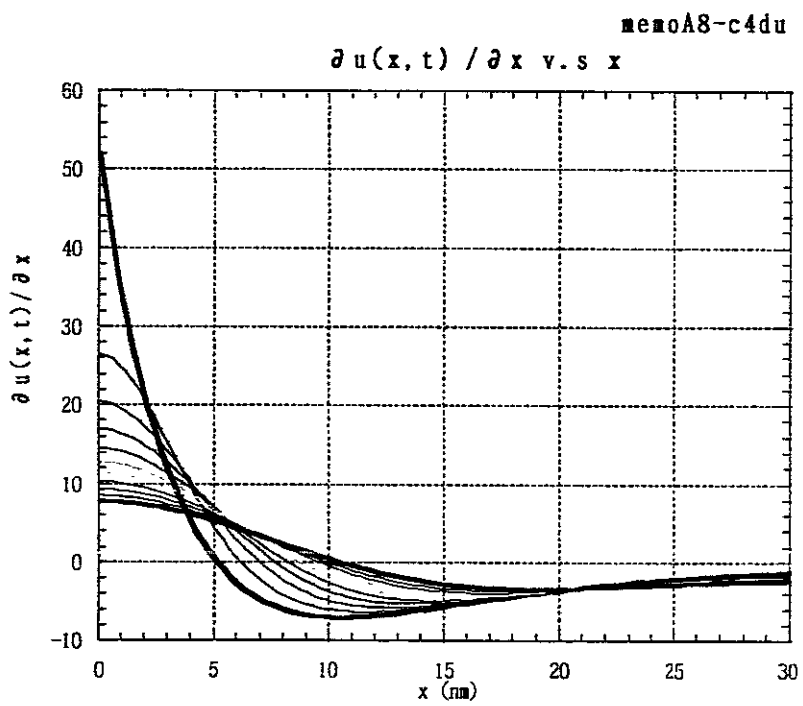


Fig. 2

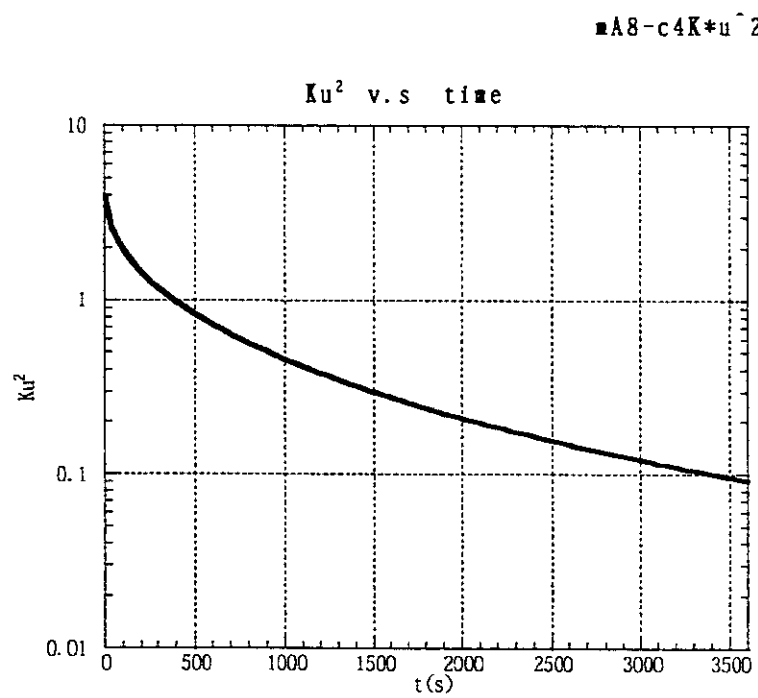


Fig. 3

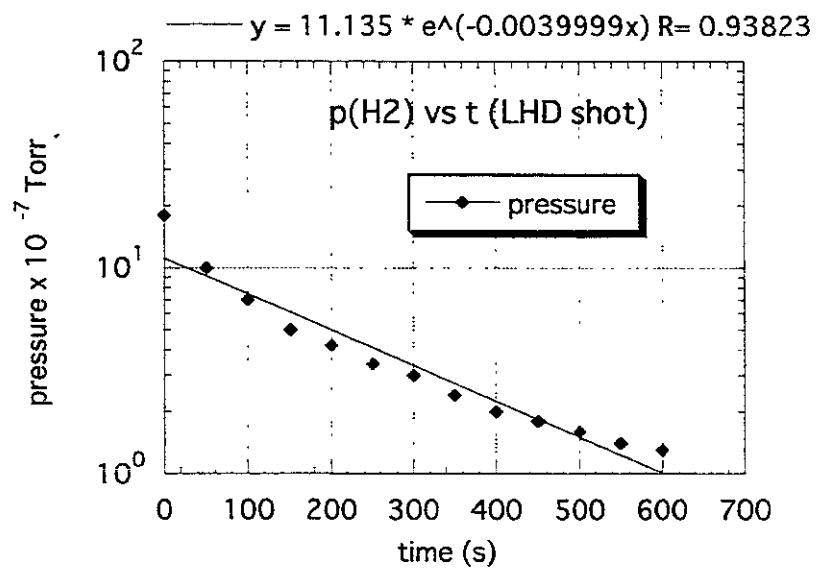


Fig. 4

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