



Investigation of Hydrogen Diffusion Profile of Different Metallic Materials for a Better Understanding of Hydrogen Embrittlement

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Highlights

- This paper focuses transient hydrogen diffusion profiles of iron, nickel, and titanium.
- Crystal structure dependence of hydrogen diffusion was investigated.
- Effect of tempering time on hydrogen back-diffusion was discussed.

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Abstract

In this study, hydrogen diffusion profiles of different metallic materials were investigated. To model hydrogen diffusion, 1D and 2D mass diffusion models were prepared in MATLAB. Iron, nickel and titanium were selected as a material of choice to represent body-centered cubic, face-centered cubic, and hexagonal closed packed crystal structures, respectively. In addition, hydrogen back diffusion profiles were also modeled after certain baking times. Current results reveal that hydrogen diffusion depth depends on the microstructure, energy barrier model, temperature, and charging time. In addition, baking can help for back diffusion of hydrogen and can be utilized as hydrogen embrittlement prevention method. Since hydrogen diffusion is very crucial step to understand and evaluate hydrogen embrittlement, current set of results constitutes an important guideline for hydrogen diffusion calculations and ideal baking time for hydrogen back diffusion for different materials. Furthermore, these results can be used to evaluate hydrogen content inside the material over expensive and hard to find experimental facilities such as, thermal desorption spectroscopy.

1. INTRODUCTION

Hydrogen embrittlement (HE) is a severe material degradation problem, which deteriorates the mechanical properties of structural materials in contact with pressurized hydrogen [1,2]. Especially, as the strength of material increases, its susceptibility to the HE also increases. Therefore, in order to use hydrogen as a clean energy source over fossil fuels safely, to develop advanced structural materials for hydrogen-related infrastructures and to utilize high strength materials in critical industries, such as defense and aviation without sacrificing the integrity and safety of containment structures, the exact mechanism of HE needs to be clarified.

Even though there are large amount of studies carried out to investigate the nature of HE since mid-eighties, the exact mechanism of HE is still unclear [3]. Yet, several candidates HE mechanisms have been suggested in the literature to uncover the interactions between atomic hydrogen and crystal defects, including stress induced hydride formation and cleavage, hydrogen induced decohesion, and hydrogen enhanced localized plasticity (HELP). Among all proposed mechanisms HELP, based on both experimental observations and theoretical calculation, is the most widely used HE mechanism [4]. HELP mechanism states that, hydrogen solutes shield the elastic interactions between dislocations and obstacles and increases the mobility of dislocations [5]. In addition, there is no one universal HE mechanism for all materials, instead each HE has different roles on different materials as well as coexistence of different HE mechanisms at the same time is also possible [5–8].

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The susceptibility of metal materials to hydrogen embrittlement (HE) is closely related to the diffusivity of hydrogen within the crystal lattice. Close-packed structures, such as austenite and deformation-induced ϵ -martensite, generally have a lower diffusivity than less dense materials like ferrites. The diffusivity of hydrogen in different crystal structures can be calculated using ab-initio simulations. In first-principle studies, it has been found that the diffusion coefficient of hydrogen in three different crystals of the iron structure can be ranked as $D_{\alpha\text{-Fe}} > D_{\gamma\text{-Fe}} > D_{\epsilon\text{-Fe}}$ [9,10].

Research using ab-initio simulations has shown that in body-centered cubic (bcc) iron, hydrogen atoms primarily occupy tetrahedral sites within the crystal structure. However, in areas of high hydrogen concentration, it has been observed that hydrogen atoms also occupy octahedral sites [11,12]. In bcc iron, it is typical for diffusion to occur through the migration of hydrogen atoms from one tetrahedral site to the nearest tetrahedral site. This migration process is known as classical over-barrier jump migration and it is the main mechanism for diffusion of hydrogen within the lattice [9–13]. In face-centered cubic (fcc) nickel, octahedral sites are the preferred location for hydrogen atoms to occupy. However, diffusion of hydrogen in fcc nickel also occurs through an indirect process, in which hydrogen atoms migrate through a metastable tetrahedral site located between two octahedral sites [13,14]. Theoretical studies on the preferred sites for hydrogen atoms in hexagonal close-packed (hcp) titanium have shown that octahedral sites are more stable. However, previous experimental work has suggested that tetrahedral sites may be more stable [15]. To fully understand the diffusion of hydrogen in hcp titanium it is necessary to take into account the different energy barriers associated with hydrogen migration between octahedral-octahedral (O-O), tetrahedral-tetrahedral (T-T) and octahedral-tetrahedral (O-T) sites [16]. Investigating the hydrogen diffusion in these three crystal structures (bcc, fcc, and hcp) is crucial for understanding the susceptibility of materials and alloys that include these crystal types to hydrogen embrittlement. Additionally, many experimental studies have been conducted to understand the effect of hydrogen on materials by introducing it into the material microstructure through pressurized gaseous hydrogen or cathodic charging of specimens. In cathodic hydrogen charging, both the specimen and an anode are submerged in an electrolytic solution, where H^+ ions accumulate on the specimen surface due to the flux produced by the potential applied to the system. [17]. Cathodic hydrogen charging is a widely used method to evaluate the effect of hydrogen on the mechanical properties of materials. By introducing hydrogen into the material microstructure through cathodic charging, researchers can study the effect of hydrogen on properties such as tensile strength, ductility, fracture toughness, and impact properties. [18–21]. However, it is important to understand how the hydrogen content and distribution in the specimens changes with charging time, temperature, and hydrogen diffusivity of the materials. Modeling the hydrogen concentration and distribution profile in the materials based on these parameters is crucial to understand the effect of hydrogen during experimental testing and evaluate the susceptibility of materials to hydrogen embrittlement. This can provide important information about how different parameters such as charging time, temperature, and hydrogen diffusivity affect the hydrogen content and distribution in the material and how it can affect the material's properties and susceptibility to hydrogen embrittlement. Additionally, hydrogen can be removed from the material and the risk of hydrogen embrittlement can be reduced by baking the specimens at specific temperatures and times. [22,23]. By modeling the concentration profile of hydrogen-charged materials after a discharge, it is possible to determine the necessary baking times and temperatures to prevent hydrogen embrittlement (HE).

This study aimed to investigate the diffusion of hydrogen in three different crystal structures of bcc iron, fcc nickel and hcp titanium. Using numerical models in MATLAB, the study simulated 1D and 2D transient diffusion profiles of hydrogen in these materials at different temperatures, charging times and baking times. Diffusion coefficients for each material were used in the model, and hydrogen concentrations were observed at different stages of the process. The study also looked into the back-diffusion of hydrogen from the material surfaces, and how it was affected by temperature and baking times. The goal of the study is to understand how hydrogen diffuses in these materials, how hydrogen concentration change with different parameters and how it affects the susceptibility of these materials to hydrogen embrittlement.

2. MATERIAL METHOD

In this study, the hydrogen diffusion model for three material types (bcc, fcc, and hcp) was defined by using Fick's second law of diffusion and numerical diffusion equations for 1D and 2D mediums, respectively. The diffusion analysis was performed using MATLAB for both 1D and 2D diffusion models. The models were used to represent the bcc, fcc, and hcp crystal structures of iron, nickel, and titanium, respectively.

Fick's second law of diffusion states that the accumulation rate of a substance in a given volume is proportional to the second derivative of its concentration with respect to position;

$$\frac{\partial C}{\partial t} = D \left(\frac{\partial^2 C}{\partial x^2} \right) \quad (1)$$

where D represents the diffusion coefficient of the specific material. The solution of the equation for a semi-infinite medium, with constant source C_s at the surface, and initial concentration C_0 at position x, can be found by using the error function. The error function is a mathematical function that describes the probability of a certain value in a normal distribution, it is commonly used in solving diffusion equations to find the distribution of concentration in the material at different positions and times

$$\frac{C(x)-C_s}{C_s-C_0} = 1 - \operatorname{erf} \left(\frac{x}{2\sqrt{Dt}} \right) . \quad (2)$$

According to Arrhenius equation the diffusion coefficient of a material can be described as;

$$D = D_0 \exp \left[- \frac{E_a}{k_B T} \right] \quad (3)$$

where D_0 is pre-exponential factor and E_a is the activation energy of the hydrogen diffusion. The diffusion coefficient of hydrogen in a single crystal can also be defined using the transition state theory [9,10];

$$D = nL^2 \frac{k_B T}{h} \exp \left[- \frac{E_a}{k_B T} \right] \quad (4)$$

where the n is a numerical coefficient, L is the distance between the two neighboring stable position of hydrogen in the crystal, for example, in bcc crystals, this would be the distance between the nearest tetrahedral sites where hydrogen atoms can reside, and h is the Planck's constant. In this study, the diffusion coefficients used for the three types of materials at different temperatures are presented in Table 1 to allow for comparison of the diffusion rates for each material at various temperatures [24].

Table 1. Diffusion coefficients of Fe, Ni, and Ti used in the study at 25 °C, 80 °C, and 100 °C temperatures [24]

Material	D (m ² s ⁻¹), 25 °C	D (m ² s ⁻¹), 80 °C	D (m ² s ⁻¹), 100 °C
Iron	2.5035×10 ⁻⁹	2.9845×10 ⁻⁹	3.1925×10 ⁻⁹
Nickel	9.3296×10 ⁻¹³	2.0007×10 ⁻¹²	2.6342×10 ⁻¹²
Titanium	4.2183×10 ⁻¹³	8.7690×10 ⁻¹³	1.1371×10 ⁻¹²

In the 1D diffusion model (Equation (2)), the concentration of hydrogen in different materials was determined using equations and diffusion coefficients at two different temperatures (25°C and 80°C). The concentration at a depth of 5 mm was calculated after specific charging times (1, 8, 24, and 72 hours) and the increments on the x-axis were set at 0.1 mm to determine the concentration at each point based on the charging time.

After that, a numerical model was used to study hydrogen diffusion in a two-dimensional medium of 10mm x 10mm. The model, referred to as transient numerical diffusion model, quantifies the hydrogen concentration $C_{x,y,t}$ in a 2D cartesian coordinate system, at a position x, y and at a specific time t;

$$\frac{C_{x,y,t}}{\partial t} = D \left[\frac{\partial^2 C_{x,y,t}}{\partial^2 x^2} + \frac{\partial^2 C_{x,y,t}}{\partial^2 y^2} \right]. \quad (5)$$

The model also allows to find the hydrogen concentration at a specific position after a time increment by;

$$C_{x,y,t+1} = C_{x,y,t} + d \left[C_{x+1,y,t} + C_{x-1,y,t} + C_{x,y+1,t} + C_{x,y-1,t} - 4C_{x,y,t} \right] \quad (6)$$

where the d is defined as;

$$d = D \frac{\Delta t}{\Delta x^2}. \quad (7)$$

The 2D transient model was used to calculate the concentration of hydrogen in a 10mm x 10mm medium at different time intervals using Equations (6) and (7). The initial hydrogen concentration was set to 0 ($C_0 = 0$) and the surface concentration was set to 1 ($C_s = 1$). The grid was created with a 0.1mm distance between nodes in both directions and the hydrogen concentration was calculated at each node with a 1-second time increment using Equation (6).

Finally, the model was used to simulate the back diffusion of hydrogen at a temperature of 100°C for various durations after 24 hours of charging for each material type. The simulation was performed with considering no hydrogen concentration on the surfaces.

3. THE RESEARCH FINDINGS AND DISCUSSION

The Figure 1 shows the 1D hydrogen diffusion profiles of iron, nickel, and titanium in a depth of 5mm for different charging times. The results indicate that hydrogen can diffuse through the entire depth in bcc Fe for all charging times because of its high diffusion coefficient in bcc structures (Figure 1a). In contrast, hydrogen can only diffuse up to 2mm and 1.2mm in nickel (fcc) and titanium (hcp) lattices respectively. These results are in agreement with previous literature, which states that hydrogen diffusion in bcc is greater than in fcc and hcp [3,25]. Additionally, it was observed that hydrogen diffusion through the lattice was accelerated with an increase in temperature (Figure 1d-1f). This is attributed to the fact that as the temperature increases, the Gibbs free energy of the system decreases and the chemical potential rate and kinetic energy of the system increases, all of which leads to an increase in hydrogen diffusion [26].

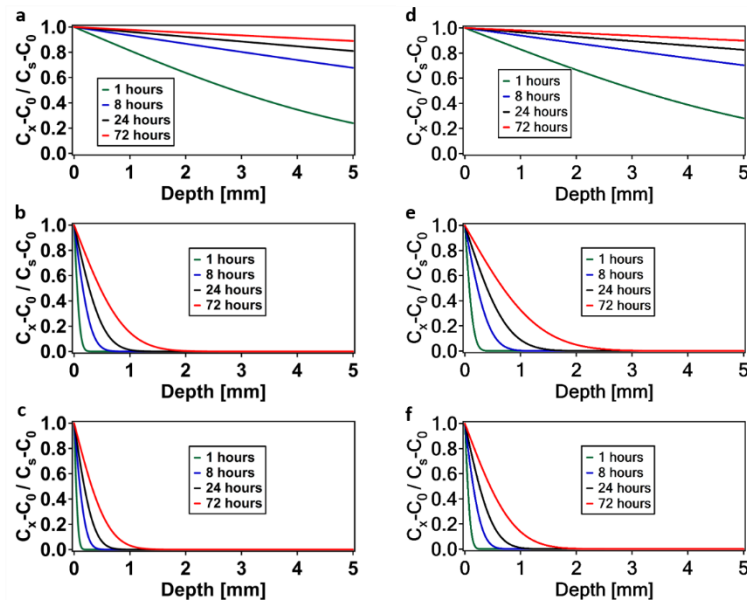


Figure 1. 1D diffusion profiles of the a) Fe b) Ni c) Ti at 25 °C and d) Fe e) Ni f) Ti at 80 °C for 1, 8, 16 and, 72 hours charging times. The Figure was retrieved from Ref. [27]

The Figure 2 illustrates the 2D diffusion profiles of hydrogen in the materials. The natural symmetry of the system is considered and only the diffusion through the x-axis is shown at $y = 5$. Compared to the 1D diffusion model, the bcc iron showed a larger difference in the amount of hydrogen diffused into the system. The concentration gap between the 1D and 2D models was more pronounced in the bcc iron due to its higher diffusion coefficient, which allows hydrogen to diffuse through the y-axis and reach 5mm in all charging times. Also, similar to the 1D diffusion model, temperature increase also enhanced the diffusion of hydrogen. It is well-known that a homogeneous distribution of hydrogen can be achieved when it reaches up to half the thickness of the specimen [7]. In conclusion, this 2D diffusion model can be used by experimentalists in electrochemical hydrogen charging setups to ensure a homogeneous distribution of hydrogen in the matrix.

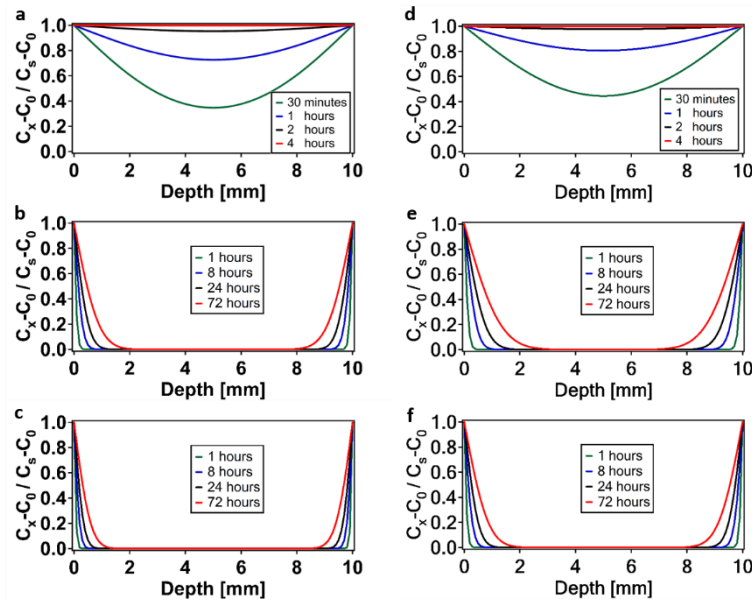


Figure 2. 2D diffusion profile of the hydrogen along x direction ($y=5$) into a) Fe b) Ni and c) Ti at 25 °C and d) Fe e) Ni and f) Ti at 80 °C. The Figure was retrieved from Ref. [27]

The results obtained from both 1D and 2D models indicate that the concentration profile of hydrogen after charging is dependent on the material type, temperature, and charging time. The results show that in bcc

iron, hydrogen concentration in the material was found to be in equilibrium with the surface concentration after 2 hours of charging. However, fcc nickel and hcp titanium lattices showed that even after 72 hours of charging, no hydrogen concentration could be observed in the center of the specimen, and the hydrogen permeation was limited to a 2mm depth subsurface region. This indicates that the crystal structure type is the most crucial parameter in determining the diffusion of hydrogen. Materials with bcc crystal structure type are found to be the most susceptible to hydrogen diffusion. In an experimental study on the diffusion profile of a β -titanium (bcc) alloy, it was observed that after 10 minutes of charging at 500 °C, the hydrogen concentration was found to be in equilibrium with the surface along a 1mm depth [28]. The effect of hydrogen on the microstructure can also be observed by the transition in the failure mode, such as a change from ductile to brittle failure of materials. Therefore, the investigation of the fracture surfaces of hydrogen-charged specimens can indicate the regions in the microstructure where hydrogen has diffused. An experimental study on a cathodic hydrogen-charged fcc nickel-based alloy found that a brittle zone of 135 μm and 300 μm depth was observed after 1 hour and 5 hours of hydrogen charging, respectively [29]. Another widely used method to understand the hydrogen accumulation within materials is Thermal Desorption Spectroscopy (TDS). TDS is a useful method that can be used to determine the amount of hydrogen in the material by measuring the amount of hydrogen that is released when the temperature of the sample is increased. Hydrogen atoms can reside in interstitial positions or can be trapped in defect sites such as grain boundaries, dislocations, and vacancies with different binding energies. The TDS method can be used to observe the hydrogen desorption flux through the surface of the material by increasing the temperature. The flux of hydrogen desorbed from the material shows a peak when the trapped hydrogen inside the defect has enough energy to overcome the binding energy [30]. An experimental TDS analysis was conducted to investigate the hydrogen desorption content of pure iron and a nickel alloy under tensile loading. The TDS results showed that the highest amount of hydrogen desorption was observed in bcc iron due to its higher diffusion coefficient [31]. This is consistent with the results obtained from the 1D and 2D diffusion models, which showed that bcc iron has a higher susceptibility to hydrogen diffusion when compared to other materials such as fcc nickel and hcp titanium.

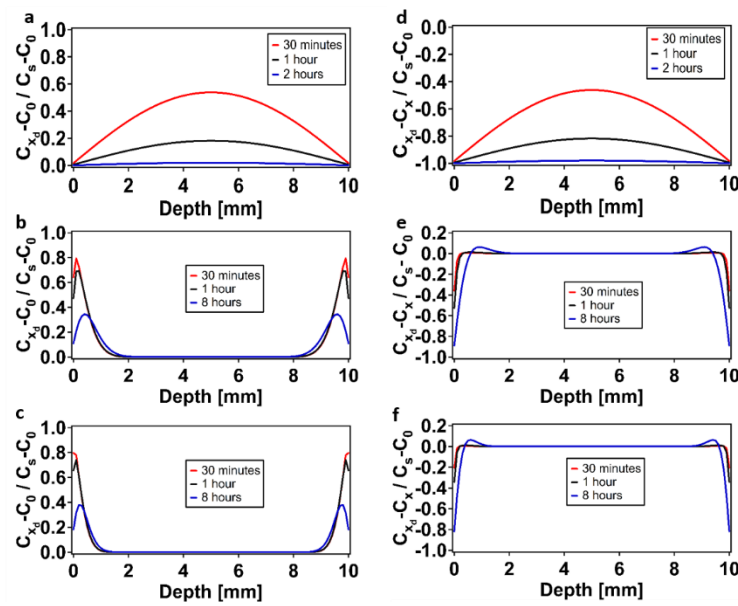


Figure 3. Remaining hydrogen concentration after the back diffusion for a) Fe b) Ni c) Ti and normalized hydrogen concentration with initial hydrogen concentration for d) Fe e) Ni and f) Ti. The Figure was retrieved from Ref. [27]

One method to prevent the mechanical degradation caused by hydrogen is to remove it from the surface by increasing the temperature. However, to specify the ideal time and temperature for back diffusion, it is necessary to understand the hydrogen content in the material and the diffusivity of hydrogen. The results of the simulations for hydrogen back diffusion in materials that were charged for 24 hours are shown in Figure 3. The remaining hydrogen concentration after the baking operations and the change in hydrogen concentration after back diffusion under 100 °C temperature were illustrated for different baking times. The results showed that most of the hydrogen concentration was accumulated in the middle of the hydrogen-charged region. Since the hydrogen concentration was in equilibrium with the surface concentration in bcc iron, the peak hydrogen content and the least change in hydrogen concentration were observed in the middle of the simulation area. On the other hand, after baking, the mass center of the hydrogen concentration was located at the subsurface for nickel and titanium lattice. During back-diffusion hydrogen was observed to permeate both through surface and through center of the medium. Thus, although fcc and hcp structures have a lower hydrogen diffusivity compared to bcc, back-diffusion of hydrogen requires longer charging times and higher baking temperatures.

4. CONCLUSION

In summary, this study investigated the 1D and 2D transient hydrogen diffusion profiles of iron, nickel, and titanium crystals that have bcc, fcc, and hcp crystal structures respectively. The results showed that the hydrogen concentration is dependent on the crystal structure, temperature, and hydrogen exposure time of the materials. The back-diffusion of hydrogen was modeled for each material under a specific baking temperature. The results indicated that although the hydrogen diffusion susceptibility of bcc crystals is higher compared to fcc and hcp structures, back-diffusion of hydrogen in these crystals requires longer baking times and higher temperatures.

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CONFLICTS OF INTEREST

No conflict of interest was declared by the authors.

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