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Development of a statistical computational model for the hydrogen parameters in high entropy alloys

Abstract

High entropy alloys (HEA's) have been the subject of intense study in recent years, due to the practical interest in it using for transporting and storage of hydrogen. The hydrogen (H) diffusion and storage parameters in these alloys are very important. In the developed statistical computational model (SCM) the coefficient of hydrogen diffusion in metals has been calculated from provisions of statistical physics. Calculations of HEA's storage parameters have been estimated with the linear regression formula. It has been found that Ta and W to increase the density of alloys and volumetric density of hydrogen in the HEA's. Vanadium in the high-entropy TiCrMnFeNi alloy was shown to provide higher hydrogen density values than zirconium with a calculated optimal concentration of 3-6 at%. The SCM model allows us to predict the values of hydrogen parameters in HEA's.

Keywords: hydrogen, computational model, high entropy alloys, diffusion coefficient, activation energy, volumetric density

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Introduction

Currently, hydrogen is used as a clean energy carrier for renewable and sustainable energy. However, the main drawback for the using of hydrogen is its efficient storage and sustainable transportation.[1,2]

The high entropy alloys have been the subject of intense study in recent years, due to the practical interest in it using for several technological applications.[3,4]

The hydrogen parameters in these alloys are very important because it leads to engineering problems associated with their creation and use for transporting and storage of hydrogen.[3–10]

In lattices, depending on the type of lattice and temperature, hydrogen can occupy two types of interstitial sites: octahedral and tetrahedral.[7–10] Even in a pure metal, transitions between interstitials of different types are possible, which entails the ambiguity of hydrogen diffusion parameters.[10,11]

First principles calculations and classical approaches, such as molecular dynamics and DFT- calculations, have been successfully used to study hydrogen diffusion in the case of body-centred cubic (BCC) metals in several papers.[14-21]

For investigation of an activation energy of hydrogen diffusion in the metals, the concept of machine learning potential (MLP) has developed.[14-16] In work,[17] the diffusivity of dilute hydrogen in bcc metals is investigated through path integral (PI) simulations using a modern MLP known as moment tensor potential (MTP).[18] These methods show a growth of the energy potentials values of H diffusion with increasing temperature, but give calculation data mainly up to 1000 K.

Thus, modern machine learning methods using first-principles calculations will provide a good description of the energy parameters in Me-H systems. However, to our knowledge, the issue of hydrogen diffusion coefficients and its storage parameters in high-entropy alloys has been insufficiently studied. Providing such calculations is important for solving practical problems of hydrogen storage in complex alloys.

In our work, we calculate the parameters of H diffusion in high-entropy alloys, as well as volumetric density of hydrogen in these alloys using the statistical computational model.

The purpose of this work is development of statistical computational model for the hydrogen diffusion and storage parameters in high entropy alloys.

Methods

Calculation of diffusion coefficient in metals

In [11] it was proposed to use statistical thermodynamic model expressions for the pre-exponential coefficient. This approach, based on the principles of statistical thermodynamics, makes it possible to adequately consider the statistical distribution of impurity atoms according to their energy for over-barrier diffusion of impurity elements. In this case, the diffusion coefficient for impurity atoms in metals is equal to[11]:

$$D = D_0 e^{-Ea/kT} = \frac{2\pi N_e Mem_a k^2 T^2 \Delta}{h^3 N \rho} e^{-Ea/kT}, \quad (1)$$

where N is the Avogadro number,

N_e is the atomic number of the element (1 for H),

M_e is the mass of 1 mole of the element ($Me = N_e$ in g/mol);

m_a is the atomic mass unit (H), ρ is the metal density,

E_a is the activation energy of diffusion,

k is the Boltzmann constant, h is the Plank constant,

Δ is the jump length of hydrogen atoms between two stable positions,

T is a temperature of diffusion.

In the developed statistical thermodynamics model the pre-exponential factor (D_0) of hydrogen diffusion in metal can be calculated by:

$$D_0 = 0.114 \cdot \frac{\Delta}{\rho} \cdot T^2 m^2 / s. \quad (2)$$

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where Δ in (2) is measured in meters, ρ is measured in kg/m³, the constant in equation (2) has a dimension kg/(m²·K²·s).

Calculation of hydrogen diffusion coefficient and activation energy in alloys

From articles on diffusion in metals, the correlation between the activation energy of self-diffusion in metals and the heat and melting point of the metal is well known.[19-21] It is also known that the activation energy of hydrogen in metals is approximately ten times less than the activation energy of self-diffusion. Therefore, for metals for which the activation energy is still unknown, we use the correlation relationship between the activation energy of hydrogen diffusion and the melting temperature of the i- element $T^i melt$:

$$E_{ai} = K_0 k T^i melt, \quad (3)$$

k is the Boltzmann constant, K_0 is the constant coefficient (~ 2.34 for H diffusion).

The K_0 coefficient was selected from the correspondence of the activation energy calculated using formula (6) with reliable data from theoretical work[13] and experimental work[19] on the diffusion of hydrogen in metals.[22]

For practical calculations of high entropy alloys (HEA's) in the developed statistical computational model, hydrogen diffusion coefficient D_H in the alloy was calculated with the linear mixing formula:

$$D^H = D_0{}^H \exp\left(-E_a{}^H / kT\right), \quad (4)$$

where $$D_0{}^H = \Sigma_{(i=1)}^N C_i D_0 i, \quad (5)$$

$$E_a{}^H = \Sigma_{i=1}^N C_i E_{ai}, \quad (6)$$

N is the number of elements in the alloy,

C_i is i-element concentration in alloy, at.,

D_{0i} is the partial diffusion coefficient H in element i, calculated by (2),

E_{ai} is partial activation energy of H diffusion in element i from Eq. 3.

Expression (3) and calculation formulas (4) – (5) are approximate. An assessment of their difference with experimental data and model parameters of DFT calculations will be performed below.

Calculation of storage parameters of high entropy alloys

Calculations of parameters of HEA's in the developed statistical computational model have been performed with the linear regression correlation:

$$P_k = \Sigma_{i=1}^N C_i P_{ik}, \quad (7)$$

where Pk is the parameter k of high entropy alloy; P_1=d is a lattice parameter;

P_2= ρ is a density of alloy; P_3= G_H is a gravimetric density of hydrogen in alloy (calculated for H/Me=1, taking into account some decrease in the density of metals);

P_{ik} is the partial parameter of metal i in high entropy alloy.

Volumetric density of hydrogen in alloy Vd was calculated from equation:

$$Vd = \rho G_H. \quad (8)$$

Results

The first example of calculating diffusion parameters is given for the TiVZrNbHf HEA's. In these alloys tuning the interstitial sites available to host hydrogen via the high-entropy effect might lead to higher storage capacity.[23] In TiVZrNbHf, a phase transition from BCC to BCT and finally to FCC was observed during hydrogenation, suggesting that the high accumulation of lattice strain due to atomic size differences in this HEM leads to a larger spectrum of interstitial sites for hydrogen absorption and to the formation of a BCT structure[24] (Alloy No 4 in Table 1).

The composition of the high entropy alloys is given in Table 1,[24] and the calculated values of hydrogen diffusion parameters are given in Table 2.

Table 1 Composition of high entropy alloys with the calculated density ρ of alloys, gravimetric G_H and volumetric density Vd hydrogen in alloys.

Cont., at. %	Ti	V	Zr	Nb	Ta	El.	Density ρ, kg/m³	G_H, %	Vd, kgH_2/m^3
1	0.225	0.225	0.33	0.22	-	-	6424.5	1.48	95.1
2	0.225	0.225	0.225	0.125	-	0.2 Mo	6981.2	1.48	103.3
3	0.25	0.25	0.125	0.25	0.125	-	7696.5	1.46	112.4
4	0.2	0.2	0.2	0.2	-	0.2 Hf	7802.2	1.33	103.8
5	0.2	0.2	0.175	0.175	0.125	0.125 Hf	8512.8	1.31	111.5
6	0.2	0.2	0.175	0.175	0.125	0.125 W	9255.3	1.31	121.3

Table 2 Calculated values for the bcc-lattice parameter d_{bcc}, the activation energy E_a and a pre-exponential factor D_0 and of H diffusivity D in HEA's.

Alloys parameters	1	2	3	4	5	6
Expt. d_{bcc}, nm [24]	0.3261	0.3181	0.3234	0.3242	0.3398	0.3393#
Calc. (SCM) d_{bcc}, nm	0.3137	0.3115	0.3138	0.3142	0.3153	0.3149
Calc. (SCM) E_a, eV	0.374	0.379	0.398	0.396	0.416	0.446
Calc. (SCM)* D_0, 10^{-8} m²/s	2.23	2.12	2.07	2	1.94	1.89
Calc. (SCM)*	1.16	0.91	0.43	0.45	0.2	0.06

*At 300 K, #Expected value

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In fact, these calculated density parameters do not consider the presence of hydrides and defects in the structure of the alloys and are of an evaluative.

It was found that Ta to increase the density and bulk density of hydrogen in high entropy alloys (No. 3 compared to No. 1 and No. 5 compared to No. 4 in Table 1) and is a promising high-entropy element.[24]

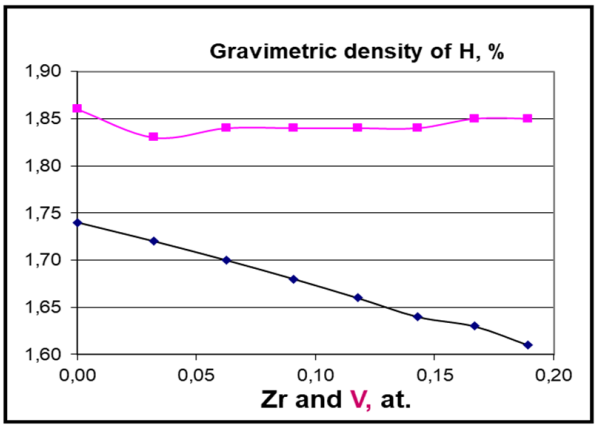
Note that alloys 3-5 presented in Table 1 may have higher hydrogen storage properties than those indicated in Table 1. Their hydrogen capacity can reach values of 2[25] and even 2.5 H/M,[23] with a hydrogen mass fraction of 2.5–2.7%. We performed calculations at the estimated saturation of these alloys' H/M=1. For comparison, the high entropy alloy $Ti_{0.3}0V_{0.25}Zr_{0.10}Nb_{0.25}Ta_{0.10}$ at the ratio H/M=1 has an experimental value of 1.35 wt.% hydrogen (Figure 7 in ref. 25), which is very close to the calculated values of alloys 1-6 in the Table 1. The addition of tungsten also increases the density of high entropy alloys, which is natural and, presumably, can increase the volumetric density of hydrogen in the high entropy alloy to a rather high value of 121.3 kgH_2/m^3. This possibility is indicated by the data of the work,[26] which shows the thermodynamic possibility of the formation of a hydrides WH and WH_4 at a sufficiently low hydrogen pressure.

We also can see that the calculated values are well correlated with the experimental values of the lattice parameter (Table 2). The experimental values are slightly higher than the values calculated based on SCM data for the range of 2.0-7.0% with a correlation coefficient R=0.92, which can be explained by the influence of H on the lattice parameter. From the correlation of calculated and experimental parameters, the expected value of the lattice parameter 0.3393 nm for a high-entropy tungsten alloy was established.

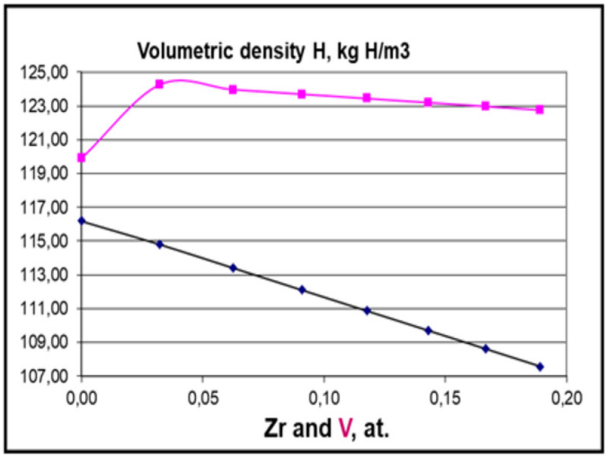
High-entropy tungsten alloy can have the lowest calculated coefficient of H diffusion, which is necessary for long-term hydrogen storage.

Another type of high-entropy hydrogen storage alloys with good reversibility and fast kinetics has been described in the C14 Laves phase.[27-29] As an example, $Ti_2Zr_XCrMnFeNi$, which contains 95 wt% of the C14 Laves phase, absorbs and desorbs 1.7 wt% of hydrogen at room temperature without activation,[28,29] comparable with the proposed hydrogen-storage material – $Ti_{1.5}V_XCrMnFeNi_{1.5}$.

From the data presented in Figure 1 we see good agreement between the calculated values of hydrogen weight density in alloys with zirconium.



a



b

Figure 1 Dependence of gravimetric and volumetric hydrogen density of HEA's from concentration of Zr and V. Blue rhombs are represented $Ti_2Zr_XCrMnFeNi$ alloy, red squares are represented $Ti_{1.5}V_XCrMnFeNi_{1.5}$ alloy.

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As is known, vanadium is an element widely used to produce high-entropy alloys and its concentration should meet certain requirements.[24,25,30,31] Vanadium in the high-entropy alloy TiCrMnFeNi provides higher hydrogen density values than zirconium at all concentrations studied. Vanadium allows us to obtain the highest hydrogen volumetric density values in the alloy $Ti_{1.5}V_XCrMnFeNi_{1.5}$ at a concentrations of 3-6 at.%.

Thus, the SCM model can be used as a basic model for preliminary calculations of hydrogen parameters in high-entropy alloys. To increase the accuracy of SCM-model calculations, should use the MLP methods for finding the H parameters in HEA's. Unfortunately, this linear regression model does not allow us to take into account all the factors of the mutual influence of elements on the parameters of hydrogen in alloys. Further development of this model will allow us to consider the influence of these factors on the parameters of hydrogen in HEA's.

Summary

For practical calculations parameters diffusion and storage of hydrogen in multi-component systems – high entropy alloys statistical computational model (SCM) has been developed. It includes first-principles calculation of the diffusion coefficient according to a statistical thermodynamic model, physicochemical calculation of activation energies for metals, and regression models of prognoses parameters in alloys.

It has been found that Ta and W to increase the density of alloys and volumetric density of hydrogen in the alloys and are the perspective elements of HEA's. The addition of tungsten can increase the volumetric density of hydrogen in the high entropy alloy to a very high value of 121.3 kgH2/m3. High-entropy tungsten alloy can have the lowest calculated coefficient of H diffusion, which is necessary for long-term hydrogen storage.

Vanadium in the high-entropy alloy TiCrMnFeNi provides higher hydrogen density values than zirconium at all concentrations studied. Vanadium allows us to obtain the highest hydrogen volumetric density at a content of 3-6 at. % in high-entropy alloy $Ti_{1.5}V_XCrMnFeNi_{1.5}$.

Thus, the SCM model can be used as a basic model for preliminary calculations of hydrogen diffusion and storage parameters in high-entropy alloys and require experimental verification.

Unfortunately, this computational model does not allow us to take into account all the factors of the mutual influence of elements on the parameters of hydrogen in alloys. Further development of this model with MLP will allow us to consider the influence of these factors on the parameters of hydrogen in HEA's.

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Conflicts of interest

None.

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