



Development of vanadium based hydrogen storage material: A review



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ABSTRACT

The metallic vanadium has an excellent hydrogen storage properties in comparison to other hydride forming metals such as titanium, uranium, and zirconium. The gravimetric storage capacity of vanadium is over 4 wt% which is even better than AB₂ and AB₅ alloys. The metallic vanadium has shown high hydrogen solubility and diffusivity at nominal temperature and pressure conditions. Consequently, vanadium is under consideration for the cost-effective hydrogen permeation membrane to replace palladium. The issues with vanadium are poor reversibility and pulverization. The poor reversibility is because of high thermal stability of β (VH/V₂H) phase which eventually restricts the cyclic hydrogen storage capacity up to 2 wt% at room temperature. The pulverization is because of large crystal misfit between the metal and metal hydride phase. The hydrogen solubility, phase stability, hydrogenation-dehydrogenation kinetics, and pulverization are highly influenced by the presence of an alloying element. Therefore, worldwide efforts are to explore and optimize the alloying element which could enhance the hydrogen solubility, destabilized the β phase, improved the hydrogenation-dehydrogenation kinetics, and prevent the pulverization. The current review is a systematic presentation of these efforts to resolve the issues of vanadium as a base material for hydrogen storage and permeation membrane.

1. Introduction

Energy supply for sustainable and cleaner environment is a never-ending challenge for the scientists and engineers. The global efforts are to restrict the global warming through minimizing the CO₂ emission by developing carbon-free and eco-friendly energy system. In connection to this the various renewable energies such as solar, wind, tidal, biomass, and sea waves are being explored in details. The prerequisite is to develop a suitable and eco-friendly energy carrier to store the renewable energies and transport with the high assurance of safety [1,2]. Hydrogen could be one of the energy carrier options because of its easy availability and eco-friendly energy conversion [2,3]. Besides the production of hydrogen using renewable energies [3], storage and transportation of hydrogen are playing a vital role to develop the hydrogen-based energy system [4–11]. Current hydrogen storage and transportation infrastructure is based on high-pressure gaseous and cryogenically cooled liquid hydrogen. The storage and transportation of hydrogen in the form of gas using high-pressure vessel are associated with several issues such as low energy density by mass, poor safety, uses of the heavy storage tank which is eventually undesirable for the mobile applications [2]. Hydrogen storage in cryogenic liquid has been

successfully employed in the space program. However, in additions to the cost of liquefaction, which amounts up to 40% of the total energy stored, use of liquid hydrogen is a concern for the public safety [3]. The hydrogen storage in the form of metal and complex hydride have been considered as a potential medium because of high hydrogen storage capacity with a high assurance of safety [11]. The hydrogen storage in metal hydride has been extensively explored and found suitable to some extent, at a desired temperature and pressure conditions [12–15]. The metal hydrides have shown a medium gravimetric storage capacity at room temperature. However; they are maintaining the superior volumetric density which is even better than the liquid hydrogen [16–21]. The desired hydrogen storage capacity as per the stipulations of United States- Department of Energy (US-DOE) is above 6 wt% [2] on material level. Although the hydrogen storage capacity of metal hydrides on a material level are below the desired level, the room temperature hydrogenation-dehydrogenation appears promising for various stationary applications. Among others, bcc vanadium based alloys have been explored extensively because of high hydrogen storage capacity (4 wt%) and easy hydrogenation-dehydrogenation kinetics at normal temperature [14,16,18,22–28]. The issues with vanadium are their poor initial activation, pulverization, and low

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cyclic hydrogen storage capacity because of the high stability of β phase [26,27]. These issues have been resolved to some extent by the addition of alloying elements [28–35]. The alloy hydrides have shown a complex thermodynamics in comparison to the pure metal hydride. The complexity had provided an opportunity to modify the nature of their interaction with hydrogen and transformed them into materials of desired hydrogen storage properties [36–40]. In the present review, the major issues and their possible solutions associated with vanadium and vanadium based alloys for hydrogen storage and permeation application have been discussed.

2. Vanadium-hydrogen system

Hydrogenation of vanadium initiated with the formation of solid solution phase which is known as α phase. In α phase, the concentration of hydrogen is directly proportional to the square root of hydrogen pressure which is known as Sieverts law as shown by Eq. (1) below:

$$C_H = K_s P^{1/2} \quad (1)$$

where, C_H is hydrogen concentration, K_s is Sievert's constant, and P is the hydrogen pressure. The Eq. (1) suggests the ingress of atomic hydrogen in vanadium matrix. Particularly, hydrogen enters as a proton (H^+) and the electrons contributed to the Fermi energy. Therefore the hydrogen solubility could be altered by changing the Fermi energy [17]. The hydrogen occupies the tetrahedral sites and will be delocalized under stress conditions. Therefore alloy with resistance to stress could alter the hydrogen solubility. The lattices parameter linearly increased with the hydrogen concentration within α phase [20]. As hydrogen concentration exceeds the terminal solid solubility (TSS), the β phase (VH/V_2H) will be nucleated. On further increase in the hydrogen concentration, the γ (VH_2) phase will be precipitated. A systematic hydrogenation process of vanadium at room temperature is illustrated in Fig. 1a–b. The first plateau appears at very low hydrogen pressure (< 1 Pa) which is corresponding to phase transformation from α to β phase. The β phase is thermally stable which required high temperature for dehydrogenation. The second plateau corresponding to the phase transformation from β to γ phase will appear near to 0.3 MPa hydrogen pressure as shown in Fig. 1a. The γ phase is unstable, and

dehydrogenation occurred at room temperature. Besides α , β , and γ hydride phases, various other phases have also been reported in the literature [28]. Interestingly, it was found that the volume increase due to hydrogenation of β phase (beyond the first plateau) is practically pressure independent [24–26]. The β is phase described as a linear VHV complexes with V-H half-bonds having a bond length of 1.76 Å [31,32]. The crystal structure of β phase is body-centered tetragonal (BCT). The crystal structure of γ phase is isomorphic with NbH_2 and possesses a CaF_2 crystal structure. The dehydrogenation pathway of vanadium hydride has been sequenced as VH_2 (γ) \rightarrow V_2H (β) \rightarrow $V(H)$ (α) \rightarrow V (H free) [33]. The decomposition temperature for these phases are 300 K, 450 K, and 750 K respectively [33,35]. The cyclic hydrogen storage capacity at a hydrogen pressure of above 0.3 MPa will be corresponding to the reaction (2) given below:



Thus, only half the total hydrogen storage capacity will be available in the subsequent hydrogenation-dehydrogenation cycles. To enhance the cyclic hydrogen storage capacity at room temperature, α , and β phases must be destabilized by tailoring the thermodynamics and kinetics by a suitable alloying element.

3. Tailoring the stability of α phase

The effect of alloying element on the stability, kinetics, and thermodynamics of hydrogenation of vanadium to form α phase have been reported [35,36]. The rate of hydrogenation in V-Ti based alloys increased noticeably with Ti content and reduced by the addition of Si, Al, and Fe [20,21]. The Ti also increases the terminal solid solubility (TSS) of hydrogen in vanadium. The V-30 at% Ti has shown TSS of hydrogen up to 0.6 H/M ratio. The terminal solid solubility (TSS) of hydrogen in vanadium increased rapidly with Nb content [38]. The Nb addition decreased the enthalpy of the V-H system reached to the maximum at 75 at% Nb in V. The enthalpy seems to be the reason of an increased TSS of hydrogen in V-Nb alloys [38]. The entropy of hydrogen in vanadium within TSS marginally increased, however, varies smoothly and continuously with niobium [28]. The Cr reduces the TSS of hydrogen in V it has very similar effect of aluminum [28].

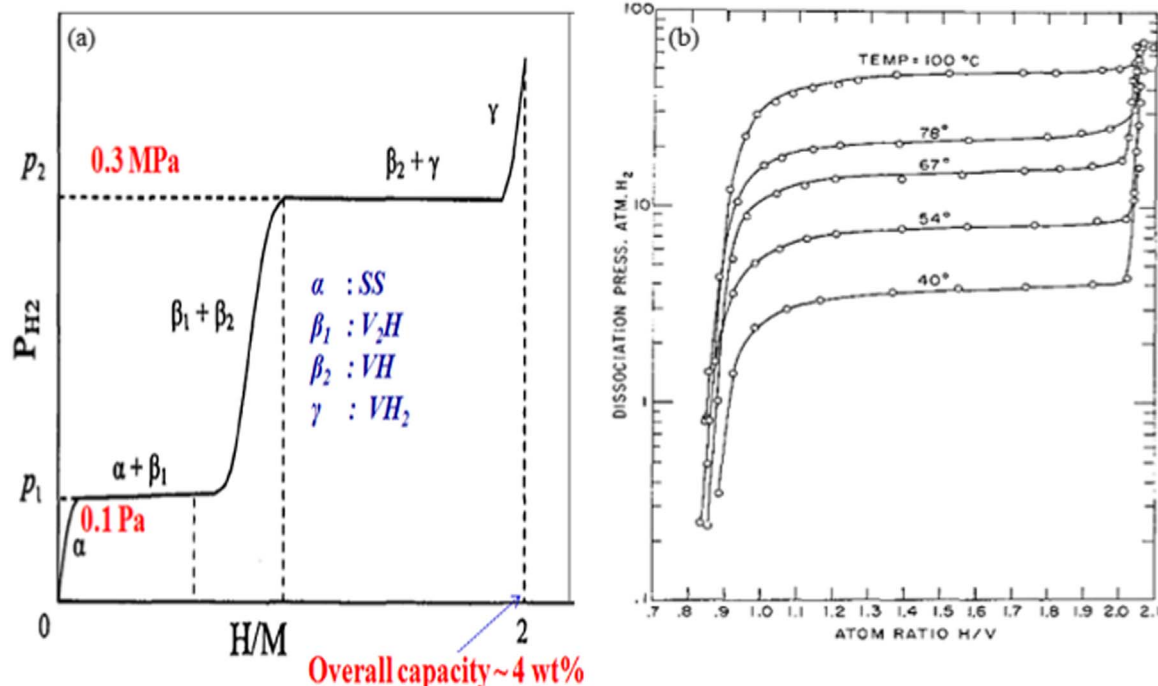


Fig. 1. Schematic of PCT curve for Vanadium metal, (b) pressure-composition isotherms for the $VH - VH_2$ transformation (Right). Reprinted from Ref. [32] with the permission of ACS.

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