

Toward room-temperature hydrogen storage with high-entropy hydrides

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Abstract Hydrogen holds great promise as a future fuel source due to its abundant availability on Earth, high energy density, and potential for clean production. When hydrogen combusts, it only produces water vapor, making it an environmentally friendly option. However, the future of hydrogen fuel hinges on efficient and safe storage methods. Efforts to achieve safe and compact hydrogen storage have focused on metal hydrides, which store hydrogen in a solid-state form. These metal hydrides offer advantages over high-pressure gas and cryogenic liquid storage, such as high volumetric density, low operational pressure, and simple hydrogen uptake. Nevertheless, a major challenge with most metal hydrides is that they require high temperatures for hydrogen desorption or activation during the hydriding process, while room-temperature hydrogen storage is essential in many applications.

High-entropy materials, containing at least five principal elements, have received significant attention in recent years for different applications. Hydrogen storage is one of these applications, but the drawback of most high-entropy alloys for this application is the need for heating. Recent experimental studies combined CALPHAD and first-principles calculations introduced several high-entropy alloys with low/room-temperature hydrogen storage and capacities higher than those of commercial materials. The main alloys designed were TiZrCrMnFeNi [1], TiZrNbFeNi [2], TiZrNbCrFe [3], $Ti_xZr_{2-x}CrMnFeNi$ ($x = 0.4-1.6$) [4], TiZrNbCrFeNi [5], TiV₂ZrCrMnFeNi [6] and TiV_{1.5}ZrCr_{0.5}MnFeNi [7] and TiV_{1.5}Zr_{1.5}CrMnFeNi [7].

Ti_xZr_{2-x}CrMnFeNi ($x = 0.4-1.6$) alloys with the Laves phase structure and low hydrogen binding energies of -0.1 to -0.15 eV were some of the best-designed and synthesized alloys. The high-entropy alloys reversibly stored hydrogen at room temperature, while pressure-composition-temperature (PCT) isotherms confirmed that (de)hydrogenation pressure was systematically reduced by increasing the zirconium fraction (Fig. 1a) in good agreement with the first principles binding energy calculations through density functional theory (Fig. 1b) as well consistent with the enthalpy predictions using the machine learning approach. The kinetics of hydrogenation was fast, the hydrogenation occurred without any activation or catalytic treatment, the hydrogen storage performance remained stable for at least 1000 cycles (Fig. 1c), and the storage capacity was higher than that for commercial LaNi₅ which is an expensive hydrogen storage material.

This talk covers some of the most significant results achieved for hydrogen storage in high-entropy alloys from the thermodynamic and kinetic aspects at Kyushu University with a focus on experimental approaches, theoretical calculations and machine learning.

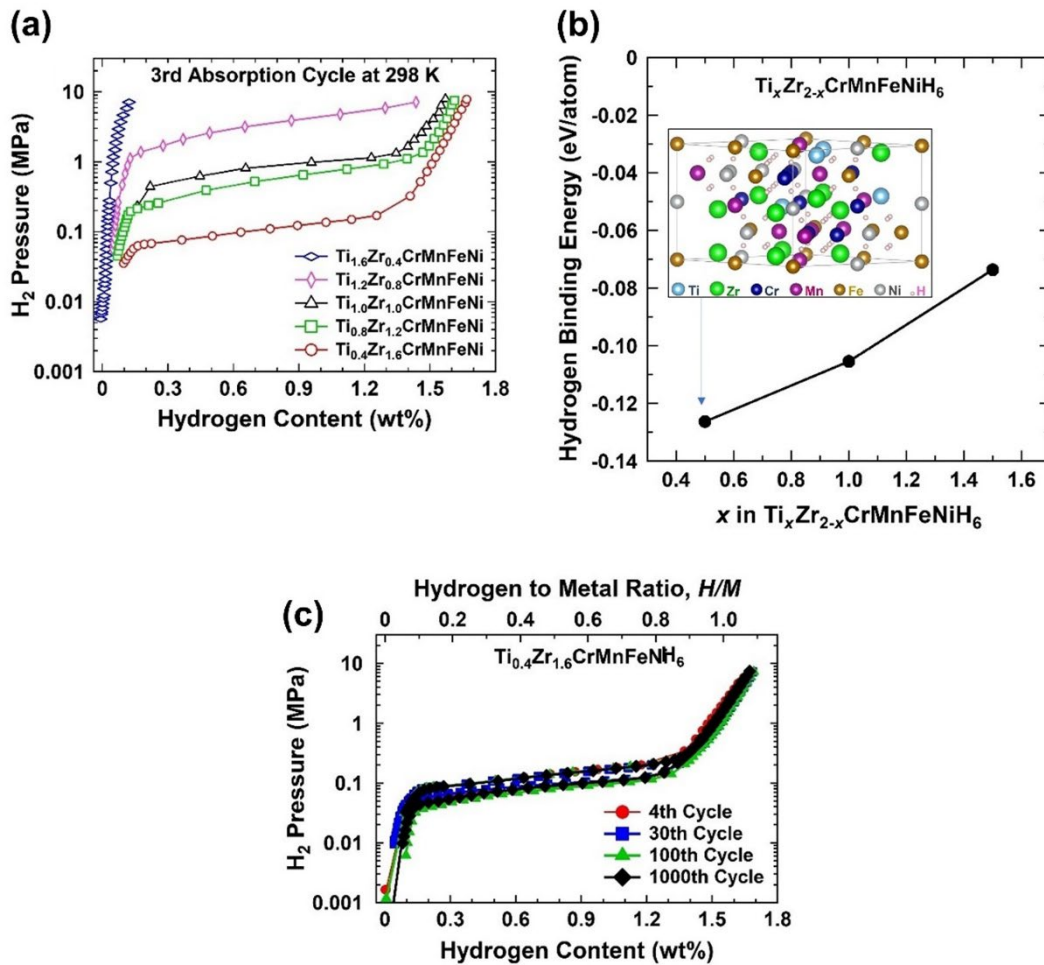


Figure 1 - (a) PCT isotherms, (b) first-principles hydrogen binding energy calculation and (c) cyclic hydrogen storage test for $Ti_xZr_{2-x}CrMnFeNi$ [4].

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