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Abstract

Hydrogen diffusion in Zr-based metallic glasses at high hydrogen contents

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Zr-based metallic glasses might be useful for hydrogen storage application: they are known to absorb high amounts of hydrogen, but exhibiting less severe embrittlement than their crystalline counterparts. In order to understand kinetics of hydrogen absorption and desorption, data on hydrogen diffusion are necessary. The aim of this paper is to analyze hydrogen diffusivities in melt-spun amorphous $Zr_{69.5}Cu_{12}Ni_{11}Al_{7.5}$ metallic glasses at high hydrogen contents.

Zr–Cu–Ni–Al metallic glasses were prepared by melt-spinning; hydrogen charging was performed electrochemically in a 2:1 glycerol–phosphoric acid electrolyte. Hydrogen desorption is hindered by a thin zirconia layer formed immediately at the surface of these Zr-based alloys. Diffusivities were measured at different temperatures by the technique of nuclear magnetic resonance (NMR) diffusion in a static fringe field of a superconducting magnet. The analysis of the echo damping allows a model-independent determination of the hydrogen diffusion constant.

For all hydrogen contents studied in this investigation, an Arrhenius-type temperature dependence of the hydrogen diffusion was observed, thus indicating a simple over-barrier-hopping mechanism. Between hydrogen contents of $H/M = 0.05$ and $H/M = 0.2$, the hydrogen diffusivity does not change; at higher contents, hydrogen diffusivity was observed to decrease until reaching a constant value at $H/M = 1.0$. The decrease of the diffusivity can be related to an increased interaction between the hydrogen atoms. At very high concentrations, there might be amorphous phase separation, thus opening new diffusion paths along interfaces and leading to a diffusivity independent of further increase of the hydrogen content.

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