MS005.O05

Local structure study during hydrogenation by time-resolved x-ray total scattering

Akihiko Machida[1], Naoyuki Maejima[1], Tetsu Watanuki[1], Hyunjeong Kim[2], Kouji Sakaki[2], Yumiko Nakamura[2]
[1]*SRRC, National Institutes For Quantum And Radiological Science And Technology, Hyogo, Japan,* [2]*Reserch Institute of Energy Frontier, National Institute of Advanced Industrial Science and Technology, Tsukuba, Japan*
E-mail: machida@spring8.or.jp

Hydrogen absorbing alloys are considered as potential hydrogen storage materials for several applications such as stationary energy storage system. The absorbed hydrogen atoms largely expand the metal lattice and induce a structural phase transition on the hydrogenation reaction. LaNi5 based intermetallic compounds are typical hydrogen absorbing materials and those crystallographic averaged structures on hydrogen absorption and desorption processes have been investigated using conventional diffraction methods [1]. Time resolved x-ray diffraction studies revealed that a transient intermediate state was formed in LaNi5 during hydrogen absorption and desorption processes [2]. We also found the formation of transient intermediate state in substituted La(Ni,Al)5 and La(Ni,Sn)5 intermetallic compounds. X-ray diffraction profiles of those compounds during hydrogenation were well reproduced by the three-phase model (alloy, hydride and intermediate). From a nanoscale structural point of view, the local structure changes would influence the formation of the intermediate state. The average structural information from conventional methods is not sufficient to elucidate the formation mechanism of the intermediate state, and therefore local structural information is also necessary.

Atomic pair distribution function (PDF), which can be obtained from the total scattering measurement, is one of potential methods to investigate the local structures. BL22XU in SPring-8, Japan enables us to perform the PDF experiments [3] by the rapid acquisition technique using the large area detector. Obtained PDFs show peaks even above r~100 Å allowing us to investigate even intermediate-r range structural features. Recently, we have developed time-resolved measurement setup using an amorphous-Si detector from PerkinElmer. This setup enables us to study change in both average and local structures of hydrogen absorbing alloys in either equilibrium or non-equilibrium states with hydrogen gas pressure up to 1 MPa.

We succeeded in measuring the time-resolved total scattering patterns during hydrogen absorption process of activated LaNi4.5Al0.5 with short accumulation time below one second. Obtained PDFs show sufficient counting statistics even at higher-r region to analyze. Some significant broadened Bragg reflection peaks are observed in the time-resolved total scattering data during hydrogen absorption, suggesting the formation of the transient intermediate state. The obtained PDF profiles also exhibit the trace of the formation of the intermediate state. The PDF profiles during hydrogen absorption cannot be reproduced by three phase model based on the averaged structure in the short-r region below ~5 Å, suggesting the intermediate state has a locally distorted structure.

This work is supported by Photon and Quantum Basic Research Coordinated Development Program from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

[1] Nakamura, Y. et al. (2000). J. Alloy. Compd. 308, 309-318.
[2] Joubert, J.-M. et al. (2006). Acta Mater. 54, 713-719.
[3] Kim, H. et al. (2013). J. Phys. Chem. C 117, 26543-26550.

Keywords: x-ray total scattering, in-situ measurement, hydrogen absorbing materials