Site occupancy of hydrogen in Ta-rich Ta-Nb alloys as observed by the channelling method†

Y. Okada,[*1, *2] E. Yagi,[*1, *2] H. Matsuba,[*2] C. Sugi,[*2] S. Koike,[*3] S. Nakamura,[*1, *2] T. Sugawara,[*4] T. Shishido,[*4] and K. Ogiwara[*1]

In metal-hydrogen systems, alloying with substitutional or interstitial solutes has significant effects on various hydrogen properties in these systems, e.g., the terminal solubility of hydrogen, hydrogen diffusivity etc. One of the important factors controlling such effects is hydrogen-solute interaction. As an example for substitutional solutes, the interaction in the Nb-based Nb-Mo and Nb-Ta alloys has hitherto been studied systematically in the solute concentration range up to 60 at. % through the observation of a change in the lattice location of hydrogen. Information on the lattice location of hydrogen had been extremely limited, because of experimental difficulties. Therefore, the channelling method utilizing a nuclear reaction of $^{1}H(^{11}B,\alpha)\alpha\alpha$ with a $^{11}B^{+}$ beam of about 2 MeV with a Tandem accelerator was developed.[1] Hydrogen can be detected by measuring emitted α particles.

Both Nb-based Nb-Mo and Nb-Ta alloy systems form a solid solution over the entire solute concentration (C_{Mo} and C_{Ta}) range, maintaining a bcc crystal structure, although the lattice parameter changes. Their atomic radii are 1.43 Å for Nb, 1.36 Å for Mo (undersized solute) and 1.44 Å for Ta (slightly oversized solute). It has been demonstrated that, in the Nb-Mo alloys, the lattice location of hydrogen changes very sensitively to C_{Mo}, whereas, in the Nb-Ta alloys, the C_{Ta} dependence of the site change is very weak. With the help of measurement of X-ray reflection line width, such a change was interpreted on the basis of lattice distortion induced by alloying.[2] Broadening of reflection lines serves as a measure of lattice distortion averaged over the whole specimen. Hydrogen is located at tetrahedral (*T*) sites in Nb and Ta.

In the Nb-Mo alloys, at low C_{Mo}, lattice is strongly distorted around individual undersized Mo atoms. Hydrogen is trapped by a Mo atom so as to reduce the lattice distortion around the Mo atom, and is located at a site displaced from a *T* site by about 0.6 Å (T_{tr} site) towards the Mo atom, indicating a strong attractive interaction between hydrogen and Mo atoms.[3] With increasing C_{Mo}, the lattice distortion is reduced owing to interference between strain fields around individual Mo atoms, and most of the H atoms occupy *T* sites as in Nb. For C_{Mo}>39 at. %, the lattice distortion gradually increases again with increasing C_{Mo}, because of an increase in the number of undersized Mo atoms in a unit cell, but not so strongly as that at low C_{Mo}, i.e., up to an intermediate level. In this case, hydrogen is distributed over *T* and *d*-*T* sites, where *d*-*T* sites are displaced from *T* sites to the nearest neighbour octahedral (*O*) sites by about 0.25 Å. Hydrogen preferentially occupies *T* sites in the undistorted or very weakly distorted region, but, as the concentration of available *T* sites is limited, excess H atoms occupy *d*-*T* sites in the region distorted at the intermediate level.[2]

In the Nb-Ta alloys, at low C_{Ta}, hydrogen is distributed over *T* and *d*-*T* sites as in the case of the Nb-Mo alloys with C_{Mo}>39 at. %. At low C_{Ta}, the lattice distortion was observed to be at the intermediate level from the measurement of X-ray reflection line width. In the 50 at. % Ta alloys, where lattice is less distorted, hydrogen is located at *T* sites. Therefore, there exists no attractive interaction between hydrogen and an oversized Ta atom.[4]

According to the above-stated interpretation, it is expected that, in the Ta-rich Ta-Nb alloys, hydrogen is distributed over *T* and *d*-*T* sites, because a size difference between Nb and Ta atoms is small, although a Nb atom is an undersized solute. In this study, therefore, the lattice location of hydrogen in the Ta-rich Ta-Nb alloy with 3 at. % Nb was investigated. This alloy is symmetrical in the solute concentration to the hitherto studied Nb-rich Nb-Ta alloys containing 2 or 5 at. % Ta atoms. The following results were obtained. Hydrogen atoms occupy *T* and *d*-*T* sites, which are displaced by 0.25–0.3 Å from *T*-sites. Hydrogen preferentially occupies *T* sites and excess H atoms enter *d*-*T* sites. This is compatible with the above-stated expectation, and is different from the case in Nb alloyed with 3 at. % undersized Mo atoms. There exists no strong attractive interaction between hydrogen and undersized Nb atoms in Ta. Considering also the results obtained for the Nb-Mo alloys, the Nb-50 at. % Ta and Nb-(2–5) at. % Ta alloys, it is concluded that, in the Nb-Mo and Nb-Ta alloys, the site occupancy of hydrogen and the interaction between hydrogen and solute atoms can be well explained on the basis of the lattice distortion induced by alloying.

References

1) E. Yagi, T. Kobayashi, S. Nakamura, Y. Fukai, and K. Watanabe: J. Phys. Soc. Jpn. **52**, 3441 (1983).
2) E. Yagi, et al.: J. Phys. Soc. Jpn. **79**, 044602 (2010).
3) E. Yagi, S. Nakamura, F. Kano, T. Kobayashi, K. Watanabe, Y. Fukai, and T. Matsumoto: Phys. Rev. B **39**, 57 (1989).
4) C. Sugi, E. Yagi, Y. Okada, S. Koike, T. Sugawara, T. Shishido and T. Ogiwara: J. Phys. Soc. Jpn. **82**, 074601 (2013).

† Condensed from the article in J. Phys. Soc. Jpn. **83** 064602 (2014).

*1 RIKEN Nishina Center
*2 School of Science and Engineering, Waseda University.
*3 Department of Physics II, Tokyo University of Science.
*4 Institute for Materials Research, Tohoku University.