

## Muon spin relaxation after hydrogen absorption-desorption process in Pd

M. Mihara,<sup>\*1,\*2</sup> H. Araki,<sup>\*3</sup> K. Shimomura,<sup>\*2,\*4</sup> W. Higemoto,<sup>\*2,\*5</sup> M. Mizuno,<sup>\*3</sup> K. Sugita,<sup>\*3</sup> Y. Tanaka,<sup>\*3</sup> Y. Kitayama,<sup>\*3</sup> D. Tomono,<sup>\*2,\*6</sup> E. Torikai,<sup>\*2,\*7</sup> W. Sato,<sup>\*8</sup> K. Ohkubo,<sup>\*3</sup> R. Murakami,<sup>\*3</sup> N. Matsuoka,<sup>\*3</sup> I. Watanabe,<sup>\*2</sup> T. Matsuzaki,<sup>\*2</sup> and R. Kadono<sup>\*2,\*4</sup>

Palladium has a unique property as a hydrogen storage material, by which it can absorb large amounts of hydrogen under ambient hydrogen gas pressure or by electrochemical charging at around room temperature.<sup>1)</sup> By using positron annihilation lifetime spectroscopy, we have found that an anomalously large amount of vacancies are formed in Pd after a hydrogen absorption-desorption process at room temperature.<sup>2,3)</sup> In addition, our recent  $\mu$ SR study on Pd has shown that in electrochemically charged Pd with hydrogen, a  $\beta$  hydride phase ( $\text{PdH}_x$ ) remains even after hydrogen desorption by degassing at room temperature.<sup>4)</sup> These results suggest that residual hydrogen atoms are possibly trapped after the desorption process due to the formation of vacancies. Although some models have been suggested, such as the formation of vacancies in a hydride phase<sup>2,3)</sup> or a vacancy-hydrogen (V-H) cluster<sup>5)</sup> in metal-hydrogen systems, no direct evidence to judge those models have been presented yet. In this report, we present the result of ZF- $\mu$ SR measurements on post-annealed  $\text{PdH}_x$  at various temperatures to clarify the microscopic property of residual hydrogen atoms in Pd.

The  $\mu$ SR experiment was performed at the RIKEN-RAL muon facility and J-PARC. The  $\text{PdH}_x$  samples were prepared by means of electrolytic charging in which a voltage was applied between Pd and Pt plates immersed in NaCl solutions. In order to minimize the time between the post-annealing and the  $\mu$ SR measurement, an equipment consisting of an electric furnace and a glass tube connected to a vacuum pump was prepared beside the  $\mu$ SR experimental port. The post-annealing was done for 1 h just before the  $\mu$ SR measurement, and the amount of removed hydrogen by the post-annealing process was deduced from the weight difference.

The ZF- $\mu$ SR spectra for each sample were well explained by a combination of a dynamic Gaussian Kubo-Toyabe (DKT) function and a nearly non-relaxing component.<sup>4)</sup> Figure 1 shows a typical result of the asymmetry of the relaxing DKT component and the

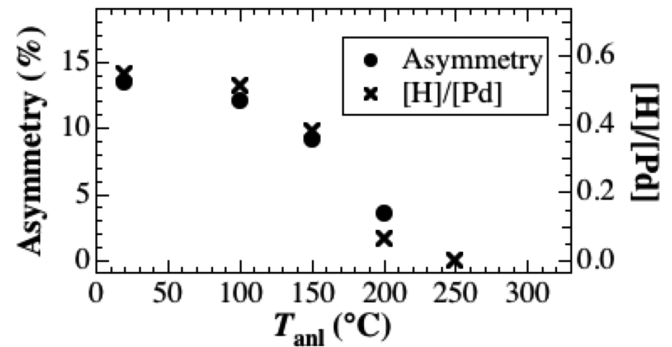


Fig. 1. Post-annealing temperature dependence of asymmetry for ZF- $\mu$ SR in  $\text{PdH}_x$  and hydrogen content.

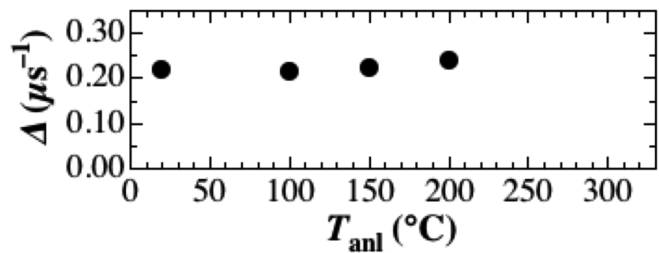


Fig. 2. Post-annealing temperature dependence of field distribution width for ZF- $\mu$ SR in  $\text{PdH}_x$ .

amount of hydrogen content in Pd as a function of the post-annealing temperature. This shows that hydrogen in  $\text{PdH}_x$  is stable up to about 100°C and is mostly removed at around 200°C, and the asymmetry follows almost the same annealing temperature dependence with the hydrogen content. The values of field distribution width  $\Delta$  for the DKT function are nearly constant and independent of the annealing temperature, as shown in Fig. 2. The present result suggests that the residual hydrogen keeps a  $\beta$  hydride phase in which hydrogen atoms stay closely together like a cluster at a trap site and are released from Pd once such a hydrogen cluster is dissociated.

\*1 Department of Physics, Osaka University

\*2 RIKEN Nishina Center

\*3 Division of Materials and Manufacturing Science, Osaka University

\*4 IMSS, KEK

\*5 JAEA

\*6 RCNP, Osaka University

\*7 University of Yamanashi

\*8 Kanazawa University

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