MLF Experimental Report	提出日 Date of Report July 24, 2012
課題番号 Project No. 2012A0089 実験課題名 Title of experiment Static/dynamic behavior in MgH_2 at high temperatures 実験責任者名 Name of principal investigator Jun Sugiyama 所属 Affiliation Toyota Central Research and Development Laboratories, Inc.	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment June 06, 2012 – June 10, 2012

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Two powder samples (MgH_2 and milled MgH_2), which were prepared at Toyota CRDL, were packed into a gold O-ring sealed titanium cell in an Ar-filled glove box. Then, the Ti cell was mounted on a graphite sample holder in order to measure wTF-, ZF- and wLF-μSR spectrum at temperatures from 300 to 650 K under high vacuum. In order to avoid an unexpected explosion at high T due to the desorption reaction, we made small holes on the window— i.e. a Ti foil with 50 μm thickness, of the Ti cell just before the measurements.

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

Following upon the μSR measurements on a hydrogen storage material, MgH_2 below 500 K, we have measured wTF-, ZF-, and wLF-μSR spectra for an “as prepared MgH_2” and “milled MgH_2” sample until 750 K, in order to study the change in μSR parameters by the H-desorption reaction. Particularly, although there are no crucial changes in crystal structure between the “as prepared MgH_2” sample and “milled MgH_2” sample, the hydrogen desorption temperature (T_d) for the latter sample decreases by about 50 K compared with T_d for the former sample [1].

Since the hydrogen desorption reaction of MgH_2 is an endothermal reaction, it was very difficult to control the sample temperature around T_d. In particular, the infrared lamp used as a heater of the present oven was found to respond too fast to stabilize the temperature around T_d. Therefore, for the milled sample, we obtained reliable data only below 570 K, whereas T_d = 620 K. Figures 1(a) and 1(b) show the temperature dependences of the relaxation rate (λ_{TF}) of the wTF-spectrum, which corresponds to a spin-spin relaxation rate, i.e., the field distribution width (Δ) due to ^{1}H nuclear field, and the field fluctuation rate (ν) for the two samples. For the as prepared sample, both λ_{TF} and ν are almost temperature independent up to 630 K, and then become very small, eventually 0 above T_d. This means

that the H-desorption reaction abrupty occures and completes at T_d. In fact, at temperatures above T_d, the wTF-spectrum exhibited a non-relaxing oscillatory signal due to the applied wTF. Furthermore, the wTF-spectrum obtained in cooling mode from 750 K did not change even below T_d, suggesting that all the hydrogen atoms were removed from the sample.

On the other hand, for the milled sample, λ_{TF} gradually decreases with temperature above 470 K, while ν apparently increases with temperature also above 470 K. This suggests that the H-desorption reaction starts to occur far below T_d (=620 K) for the milled sample. Moreover, the increase in ν implies rapid diffusion of H^+ and/or μ^+ in the sample, which is proposed to be a significant factor to decrease T_d for the other hydrogen storage materials, such as $LiAlH_4$ [2] and $M(BH_4)_n$ [2].

However, in order to further elucidate the mechanism on the decrease in T_d by milling and/or by adding a small amount of Nb_2O_5, we need more systematic measurements at high temperatures, particularly below the vicinity of T_d using a more reliable temperature control system.

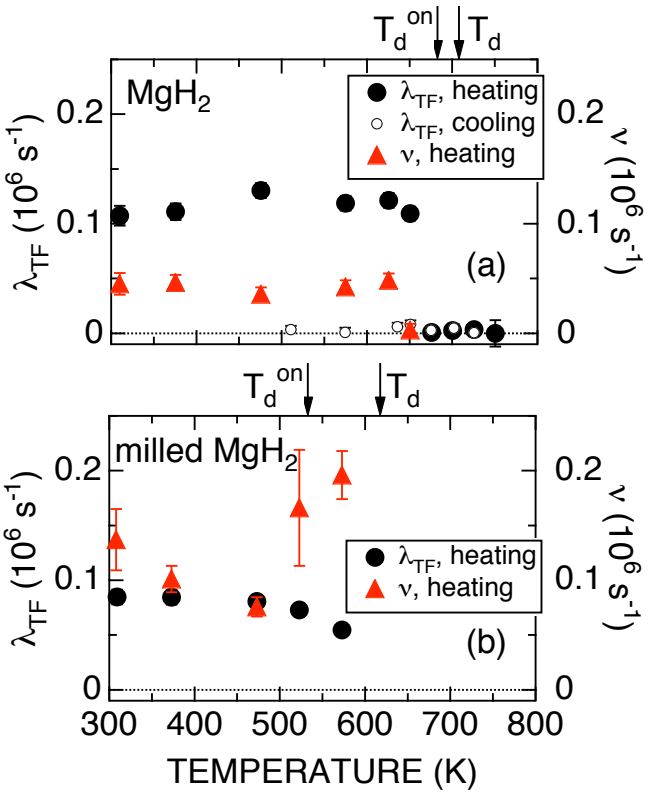

Fig. 1: The temperature dependences of the wTF relaxation rate (λ_{TF}) and the field fluctuation rate (ν) for (a) the as prepared MgH_2 and (b) milled MgH_2 sample. The data of λ_{TF} were obtained by fitting the wTF-spectrum using an exponentially relaxing cosine oscillation function, and that of ν were obtained by fitting the ZF- and wLF-spectra using a dynamic Kubo-Toyabe function. Here, wTF was 20 Oe and wLF was 10 Oe.

REFERENCES

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[3] J. Sugiyama, et al., Phys. Rev. B **81**, 092103 (2010).