MLF Experimental Report	提出日 Date of Report July XX, 2013
課題番号 Project No. 2013A0019 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 April 04, 2013 – April 07, 2013

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.

A powder sample of milled MgH_2, which was prepared at Toyota CRDL, was packed into a gold O-ring sealed titanium cell in an Ar-filled glove box. Then, the Ti cell was mounted on an improved graphite sample holder (see Fig. 1) in order to measure wTF-, ZF- and wLF-μSR spectrum at temperatures from 300 to 650 K under high vacuum. Particularly, in order to stabilize the temperature of the sample during the desorption reaction, the heat capacity of the oven was increased by about 100% than the past. Furthermore, in order to avoid an unexpected explosion at high T due to a desorption reaction of hydrogen, four small holes were made on the Ti foil window immediately 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.

According to the μSR experiment on an “as-prepared MgH_2 sample” until 750 K, both the wTF relaxation rate (λ_{TF}) and the field fluctuation rate (ν) are almost temperature independent up to 630 K, and then they become very small, eventually 0 above the hydrogen desorption temperature (T_d) (Fig. 2). This means that the H-desorption reaction abruptly occurs and completes at T_d for the “as-prepared sample”. In fact, at temperatures above T_d, the wTF-spectrum exhibited a non-relaxing oscillatory signal. Furthermore, the wTF-spectrum obtained in cooling mode from 750 K no longer changed with T even below T_d, suggesting that all the hydrogen atoms were released from the sample.

On the other hand, for the “milled sample”, λ_{TF} gradually decreases with temperature above 470 K, and then apparently increases with temperature (Fig. 2). This suggests that the H-desorption reaction starts to occur far below T_d (= 620 K) for the milled sample. Moreover, the small increase in ν implies that H^+ and/or μ^+ diffuse rapidly in the sample. Such rapid diffusion is proposed to be a significant factor to decrease T_d for the other hydrogen storage materials, such as $LiAlH_4$ [2] and $M(BH_4)_n$ [3].

2. 実験方法及び結果（つづき） Experimental method and results (continued)

For the “milled with Nb_2O_5 sample”, the behavior observed in the “milled sample” is more drastic. That is, as *T* increases from 300 K, the $\lambda_{TF}(T)$ curve shows a step-like decrease around 450 K, while ν starts to increase at 450 K and the $\nu(T)$ curve has a clear maximum around 550 K. This indicates that the doped Nb_2O_5 enhances the H-desorption reaction of the “milled sample” at low temperatures.

Then, we reattempted to measure μSR spectra for the “milled sample” using an improved oven in order to elucidate the change in the μSR parameters during the H-desorption reaction. However, due to a failure of the valve control of the vacuum system, it was unable to heat the sample up to the H-desorption temperature. In order to complete the study, we definitely need to measure the “milled sample” in the next cycle.

Fig.1 An improved oven for the MgH_2 measurements. Thanks to this oven, we obtained the change in a dynamic behavior of “milled with Nb_2O_5 sample” during the H-desorption reaction.

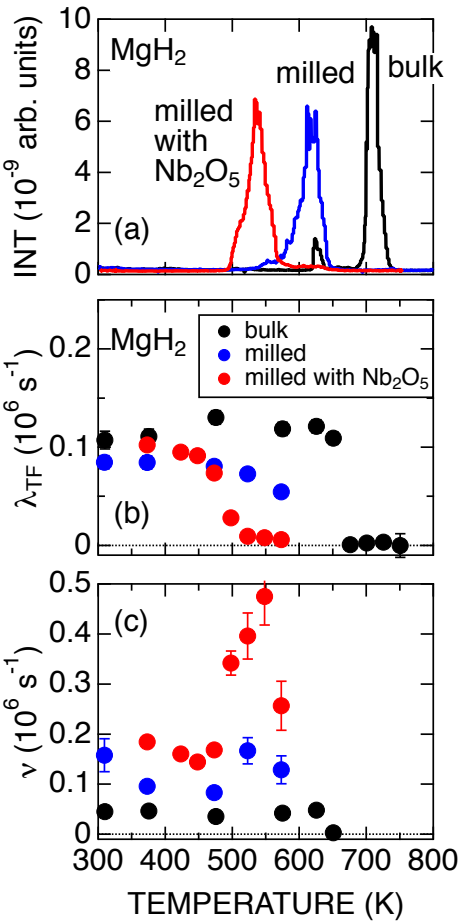

Fig. 2 The temperature dependence of (a) TPD curve, (b) wTF relaxation rate (λ_{TF}), and (c) the field fluctuation rate (ν) for the three samples. Here, λ_{TF} corresponds to the spin-spin relaxation rate, while ν to the spin-lattice relaxation rate.

REFERENCES

[1] N. Hanada, et al., J. Alloys and Compounds **446-447**, 67 (2007).

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