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## Structures and Magnetic Properties of $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$ in Amorphization Process

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We have studied structural and magnetic properties of a ternary intermetallic compound  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  in the amorphization process using a high-energy ball mill. The X-ray diffracted intensity decreased, whereas the spontaneous magnetization at 0 K increased with the milling time. In Arrott plot analyses, the samples after milling were ferromagnetic. It is suggested that a sample subjected to a ball mill is in the amorphization process and a little disorder generated by the ball milling significantly affects the magnetic properties in this ternary compound system.

**KEYWORDS:**  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$ , Laves-phase compounds, ball milling, amorphization, X-ray diffractometry, magnetization measurements

Ternary intermetallic compounds  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$ , which are the hexagonal ( $\text{MgZn}_2$ -type) Laves-phase crystals for  $0 \leq x \leq 1$ , show several magnetic structures depending on the Ta concentration  $x$ .<sup>1)</sup> Magnetic properties of an amorphous  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  obtained by vapor-quenched method are different from that of the crystal including the same atomic concentration. The amorphous  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  were ferromagnetic for the whole  $x$ .<sup>2)</sup> To investigate the structures and the magnetism in the process between the crystalline and the amorphous  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$ , we have subjected these crystalline compounds to mechanical alloying or mechanical attrition (MA) using a high-energy ball mill. It is well known that the MA of an alloy causes the amorphization of the crystal.<sup>3,4)</sup>

The ball milling was performed with a MITSUI AT-TRITOR MA01D (Mitsui Miike Eng. Corp.) with a ball-to-powder weight ratio of 20:1. This milling ball was made from  $\text{Al}_2\text{O}_3$  having a purity of 92%. A ternary compound  $\text{Hf}_{0.5}\text{Ta}_{0.5}\text{Fe}_2$  was prepared by arc melting under argon atmosphere. The arc-melted button placed in a sealed quartz ampule in pressures of  $10^{-6}$  Torr was annealed at 800°C for 142 h, crushed into powder and subjected to the ball mill. We have used the samples milled for 0 h (crystal), 6 h and 24 h. The mean particle size of these samples is about 40  $\mu\text{m}$ , 6  $\mu\text{m}$  and 3  $\mu\text{m}$  in diameter, respectively, if the particle is a circle with the same area. X-ray diffractometry and magnetization measurements were used to characterize these samples. The magnetization from 6 to 340 K in magnetic fields up to 50 kOe was measured with a SQUID magnetometer.

Figure 1 shows the X-ray diffractometry data for the crystalline  $\text{Hf}_{0.5}\text{Ta}_{0.5}\text{Fe}_2$  and the materials milled for 6 and 24 h. The pattern for the sample milled for 6 h was similar to one for the crystal. From this observation, it could be seen that these two samples had little structural difference as far as we used Cu  $K\alpha$  radiation. It was also observed that the intensities of the Bragg peaks after milling for 24 h decreased. The  $2\theta$  angles of the peaks after ball milling were not different from one of the crystal. This is an indication that the hexagonal  $\text{MgZn}_2$ -type structure of these samples is gradually destroyed by the

ball milling, but the lattice parameters do not change largely.

The magnetization isotherms obtained at 6 K up to 50 kOe are shown in Fig. 2. The magnetization in the same field increased as the milling time increased. The initial magnetization curve of the ball-milled samples was steeper than that of the crystal. In field above 5 kOe, a tendency to saturation was observed for all samples. In Fig. 3, we show the temperature dependence of the magnetization in field of 5 kOe. The magnetization of the samples after milling increased remarkably below about 200 K. Similar measurements were performed in various fields. The magnetic behaviors of the samples milled for not only 24 h but also 6 h were different from that of the

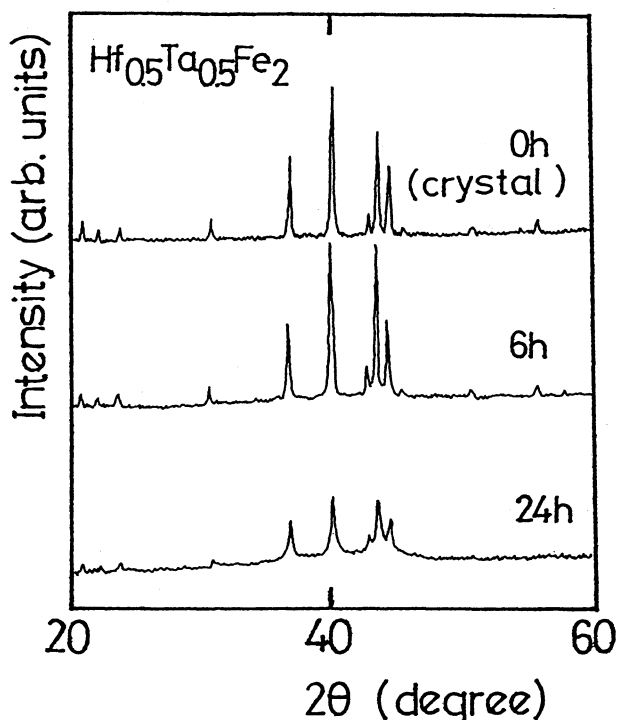


Fig. 1. X-ray diffraction patterns of  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  milled for 0 h (crystal), 6 h and 24 h.

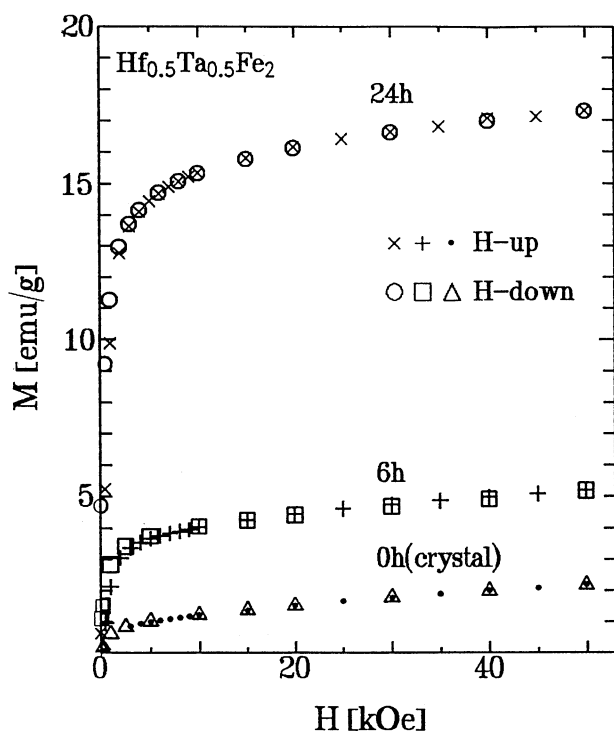


Fig. 2. Magnetization as a function of applied field up to 50 kOe for  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  milled for 0 h (crystal), 6 h and 24 h.

crystal. This suggests that the effect of a little disorder in the lattice appears not in the X-ray diffraction pattern but on the magnetic properties. In the analysis of  $M^2$  versus  $H/M$  plot (Arrott-plot) between 1 kOe and 5 kOe, it was shown that the samples after milling were ferromagnetic. The Curie temperature,  $T_c$ , of the samples milled for 6 h and 24 h was 152 K and 136 K, respectively. The spontaneous magnetization at 0 K,  $M_s(0)$ , was also 2.0 emu/g and 9.9 emu/g, respectively. It can be seen that  $M_s(0)$  increases with the milling time, and  $T_c$  decreases. The Curie temperature,  $T_c$ , and  $M_s(0)$  of the sputtered amorphous film with  $x=0.5$ , which is considered to correspond to the material subjected to a ball mill for infinite time, were about 116 K and 24.0 emu/g, respectively.<sup>2)</sup> It seems that  $T_c$  and  $M_s(0)$  of the ball-milled samples approach the values of the sputtered amorphous sample respectively. The crystalline  $\text{Hf}_{0.5}\text{Ta}_{0.5}\text{Fe}_2$  is an antiferromagnet with the Néel temperature  $T_N=268$  K.<sup>1)</sup> It is in-

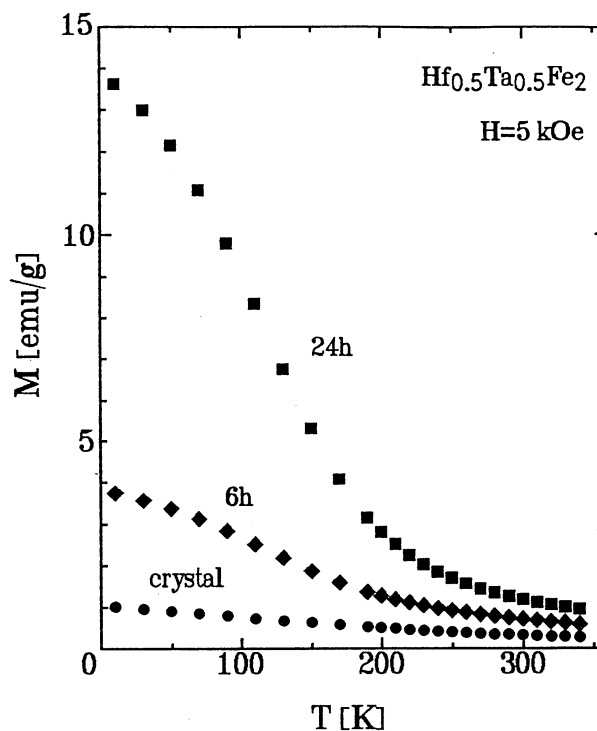


Fig. 3. Temperature dependence of the magnetization in field of 5 kOe for  $\text{Hf}_{1-x}\text{Ta}_x\text{Fe}_2$  milled for 0 h (crystal), 6 h and 24 h.

dicated that a transition from antiferromagnetic to ferromagnetic state is induced by the ball-milling process.

In conclusion, it is suggested that a sample subjected to a ball mill is in the amorphization process and a little disorder generated by the ball milling significantly affects the magnetic properties in this ternary compound system.

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