

# Amorphous 5f Ferromagnetic Hydrides $\text{UH}_3\text{Mo}_x$

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$\gamma$ -U metal, stabilized down to room temperature in the *bcc* structure by Mo doping, can absorb hydrogen at high  $\text{H}_2$  pressures only. The product is amorphous hydride  $\text{UH}_3\text{Mo}_x$  analogous to  $\beta\text{-UH}_3$ . Such hydrides are ferromagnetic with high Curie temperatures (up to 200 K), enhanced with respect to  $\beta\text{-UH}_3$ . Magnetic moment of U also increases. Large disorder together with high anisotropy lead to a very high coercivity, reaching 4 T at low temperatures. As amorphization normally tends to suppress magnetic ordering of U compounds, such hydrides represent a new class of materials, amorphous U-based ferromagnets with relatively high Curie temperature.

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## 1. Introduction

Uranium metal interacts with hydrogen, easily forming a fine pyrophoric powder of  $\beta\text{-UH}_3$ , ferromagnetic below  $\approx 170$  K. We succeeded to stabilize the *bcc* form of U (known as high temperature phase of U) by Mo doping [1]. We have been interested in reaction of these materials with H and in structure and magnetic properties of the product.

## 2. Material and methods

$\text{UMo}_x$  alloys ( $x = 0.14, 0.18, 0.22, 0.33$ ) in the *bcc* cubic structure ( $\gamma$ -phase) were prepared by arc-melting in Ar atmosphere. In addition, splat cooling was used for samples with low Mo concentrations ( $x = 0.14, 0.18$ ), which helps to retain the  $\gamma$ -phase down to the room temperature [1].

All samples were exposed to  $\text{H}_2$  gas and characterized by XRD. The products were subjected to magnetization, specific heat and resistivity measurements using the PPMS equipment.

## 3. Results and discussion

Hydrogenation attempts were performed at several  $\text{H}_2$  pressures at room temperature. Samples start to absorb hydrogen at pressures higher than 4.5 bar, but the absorption is very slow. Most of the samples were hydrogenated at pressures  $\approx 80$  bar. To quantify the amount of absorbed hydrogen, the hydride samples were decomposed in a close evacuated volume by heating to 500 °C. The total amount of hydrogen close to 3.0 H/U atoms was obtained. Therefore we denote these hydrides as  $\text{UH}_3\text{Mo}_x$ . All hydrides are quite stable in air and not pyrophoric like  $\beta\text{-UH}_3$ .

XRD analysis of all hydrides reveals broad peaks due to an amorphous structure (Fig. 1). Nevertheless, the structure can be related to diffraction lines of  $\beta\text{-UH}_3$  with

small amount of UC and  $\text{UO}_2$  impurities. A quantitative analysis by using the Rafaja model [2] shows that the experimental patterns can be reproduced assuming a broadening due to very small grains of two different sizes, namely 7–12 Å and 12–18 Å, with some variations between different Mo concentrations.

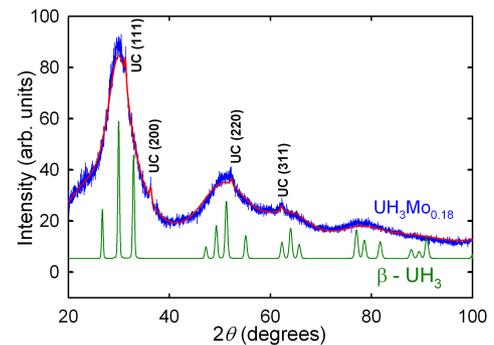


Fig. 1. XRD pattern of  $\text{UH}_3\text{Mo}_{0.18}$  (blue) compared with the simulated XRD pattern of  $\beta\text{-UH}_3$  (green) and our fit of the experimental data (red) used for the grain-size calculations.

The temperature dependence of electrical resistivity (not shown here) exhibits a dominant weak negative slope ( $d\rho/dT < 0$ ). Such negative slope was observed earlier for the U-Mo alloys with the *bcc* structure [3]. Its origin can be seen in weak localization, which appears under conditions of strong disorder, and which is partly suppressed by electron-phonon scattering. The kink observed at  $T = 200$  K is related to magnetic ordering temperature  $T_C$ . Such a value  $\approx 30$  K is higher than that for  $\beta\text{-UH}_3$  [4]. Resistivity values are unusually high, exceeding  $1000 \mu\Omega\cdot\text{cm}$ .

The temperature dependences of specific heat for hydrides with various Mo concentrations and for  $\beta\text{-UH}_3$  [5] are shown in Fig. 2. The kinks related to  $T_C$  can be observed in the range 175–200 K. The highest  $T_C$  was observed for the hydride  $\text{UH}_3\text{Mo}_{0.18}$ .  $T_C$  is reduced

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with further increase of Mo concentration. The low-temperature part of specific heat can be approximated by the Debye model, from which we estimate the Sommerfeld coefficient of the electronic specific heat. The best fit (Fig. 2 inset) was obtained with the addition of magnon contribution to specific heat  $C = \gamma T + \beta T^3 + \alpha T^{1/2} \exp(-T_0/T)$ . This fit yields  $\gamma \approx 30$  mJ/mol K<sup>2</sup>, which is in a good agreement with literature data for  $\beta\text{-UH}_3$  (29 mJ/mol K<sup>2</sup> [6]).

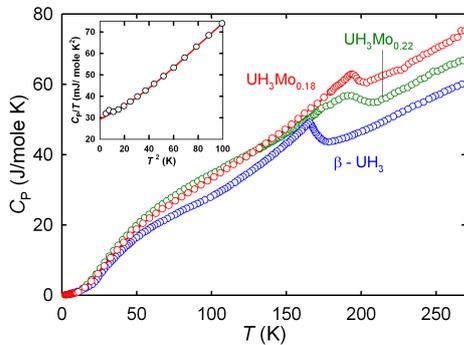


Fig. 2. Temperature dependence of specific heat of  $\text{UH}_3\text{Mo}_x$  hydrides compared with the data on  $\beta\text{-UH}_3$  [5]. The inset shows the fit of low temperature part for  $\text{UH}_3\text{Mo}_{0.18}$ .

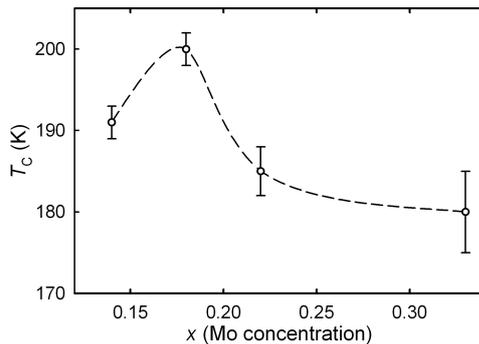


Fig. 3. Concentration dependence of Curie temperatures.

The temperature dependence of magnetization measured in various magnetic fields indicates the ferromagnetic ordering in all samples with maximum value of  $T_C$  close to 200 K for  $\text{UH}_3\text{Mo}_{0.18}$  (Fig. 3). Magnetization curves reveal similar behaviour for all hydride samples. Figure 4 displays hysteresis loops at different temperatures for  $\text{UH}_3\text{Mo}_{0.33}$ . With temperature decreasing down to 3 K the coercive field strongly increases and reaches maximum value  $\approx 4$  T. An abrupt Barkhausen jump appears at  $T = 1.7$  K between 3.5–4 T (demagnetization by shifting of domain walls), which is followed by another more smooth increase in magnetization in higher fields, associated with rotation of magnetization towards the field direction. Such behaviour can be ascribed to pinning

of domain walls in highly disordered systems with high anisotropy known as HARD materials (High Anisotropy Random Distribution). The value of spontaneous magnetic moment at  $T = 4$  K is close to  $1\mu_B/\text{U}$ , which is higher than that for  $\beta\text{-UH}_3$  ( $0.87\mu_B/\text{U}$  at 4 K [4]).

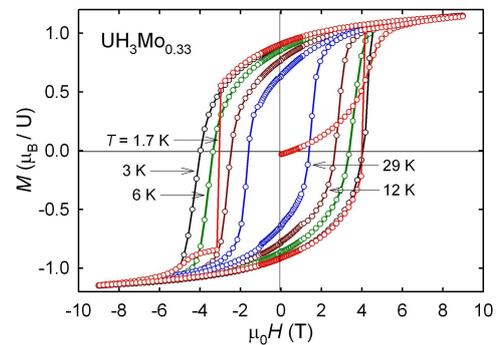


Fig. 4. Hysteresis loops of  $\text{UH}_3\text{Mo}_{0.33}$  measured at various temperatures.

#### 4. Conclusions

$\text{UH}_3\text{Mo}_x$  hydrides are quite unique 5f materials with almost amorphous structure and, at the same time, ferromagnetic ordering with relatively high  $T_C$  (30 K higher than  $\beta\text{-UH}_3$ ). That can be explained by larger U-U distances due to embedded Mo atoms. The reduced 5f-5f overlap leads to the 5f band narrowing. However, higher Mo concentrations reduce the exchange interaction between U moments, yielding the decrease of  $T_C$ . As atomic disorder leads typically to the suppression of the 5f magnetism, the new amorphous hydrides with high  $T_C$  have very exceptional position among U compounds.

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