

High pressure X-ray and Raman studies of the selected metal hydrides

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Metal hydrides, considered as promising hydrogen-storage material and potential high-temperature superconductors, have attracted increasing attention from researchers in the fields of physics and material sciences. The most efficient way to produce hydrides is the employment of high hydrogen pressure, which increases dramatically the chemical potential of hydrogen. The recent development of high-pressure diamond anvil cell (DAC) technique allowed a series of metal hydrides to be synthesized: RhH_2 , IrH_3 , FeH_3 .

This dissertation aims to seek new metal hydrides and further investigate their structural properties by means of high-pressure energy-dispersive X-ray diffraction and Raman spectroscopy. In this thesis, our studies are mainly divided to two parts. The first part focuses on high-pressure structural investigations of selected rare-earth metal hydrides. Our aim is to continue and try to complete the systematic high-pressure study on the remaining rare-earth (RE) metal hydrides. In the second part, we are devoted into seeking and synthesis of new transition metal (TM) hydrides by using high-pressure of hydrogen gas in DACs.

This thesis is composed of four chapters. Chapter 1 gives a comprehensive introduction of the studies and provides related background knowledge of the high-pressure experimental techniques employed in this thesis: high-pressure energy-dispersive X-ray diffraction and high-pressure Raman spectroscopy.

Chapter 2 presents the structural investigations on rare-earth trihydride, specifically, trihydrides of dysprosium and lanthanide. Synchrotron X-ray and Raman scattering investigations have been performed on dysprosium trihydride at high pressure up to 40 GPa. As predicted by theoretical calculation and other studies, dysprosium trihydride was observed underwent an hexagonal-to-cubic phase transformation similar to those of other heavy lanthanide trihydrides. These studies completed the overall picture of hexagonal to cubic

phase transition for the whole REH_3 compounds since dysprosium trihydride is the last one that has not been studied so far among the heavier lanthanide trihydrides family. LaH_3 and LaD_3 were chosen to be studied at high pressure due to the appearance of unknown rich peaks in our preliminary Raman measurements. The stoichiometric lanthanum trihydride and deuteride undergo pressure-induced phase transformation presumably of hydrogen ordering origin similar to that observed in the heat capacity measurements at low temperature.

Chapter 3 elucidates our recent achievements in the high-pressure investigations of the selected transition metals, molybdenum and tantalum, under high pressure of hydrogen in diamond anvil cell. The molybdenum-hydrogen system was studied in a diamond anvil cell at high hydrogen pressure up to 30 GPa at room temperature by X-ray diffraction. A phase transformation was observed of a bcc metal to a hydride with an hcp metal lattice and $H/Mo \approx 1.1$ at pressure around 4 GPa; the hydrogen content of the hydride increase with pressure increase in the pressure range of 4-15 GPa; eventually the hydrogen content reached saturation around 15 GPa with ratio H/Mo of 1.35(10) evaluated from the volumetric consideration. Although the theoretical calculation predicted the formation of dihydride we failed to obtain it. With respect to the tantalum-hydrogen system, we investigated it by x-ray diffraction in a diamond-anvil cell at room temperature in the pressure range up to 41 GPa. A substoichiometric tantalum monohydride with a distorted bcc structure was formed at 5 GPa and its hydrogen content reaches $H/Ta = 0.92(5)$; a new hydrogen-rich phase of tantalum with an hcp metal lattice was found at higher pressure and its hydrogen content was independent of pressure. The single-phase hydrogen-rich phase was synthesized at a hydrogen pressure of 9 GPa in a toroid-type high-pressure apparatus and possess an hcp metal lattice with $a = 3.224(3)$ and $c = 5.140(5) \text{ \AA}$ at $T = 85 \text{ K}$ investigated by X-ray diffraction. The hydrogen content determined by thermal desorption was $H/Ta = 2.2(1)$. The tantalum dihydride was successfully synthesized and possesses hexagonal metal lattice, unlike niobium and vanadium dihydrides which based on a fluorite structure.

In Chapter 4, a brief overview of the whole studies and final conclusions is given.