SAND2007-5844P

In-Situ Optical Diagnostics of Neutron Generator Target Films

LABORATORY DIRECTED RESEARCH & DEVELOPMENT

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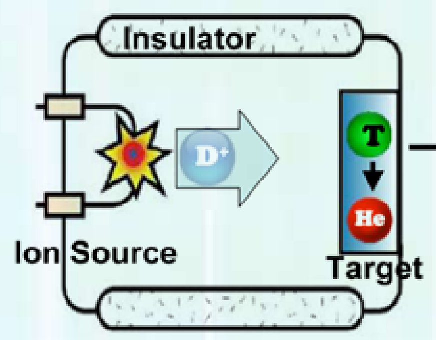
Research Problem

- Neutron tubes are small electrostatic accelerators that produce neutrons. Deuterium ions are accelerated into a target containing tritium to produce neutrons via the reaction:

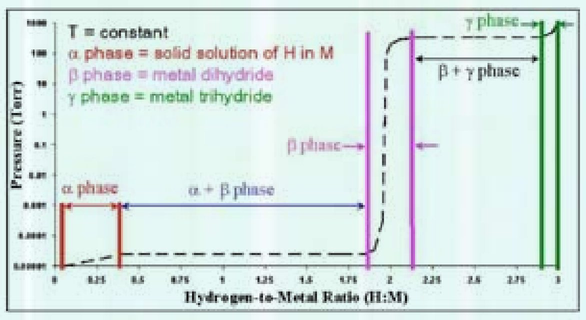
$$D^+ + T \rightarrow n^0 + He^{4+}$$

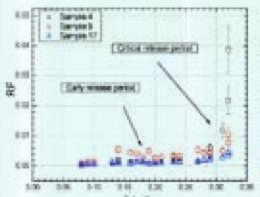
- Target and source store the hydrogen isotopes deuterium (D) and tritium (T) in metal occluder films.
- Tritium is radioactive and decays into 3He, which if released from the target film will poison the vacuum environment of the neutron tube.



Why do the occluders need special attention?

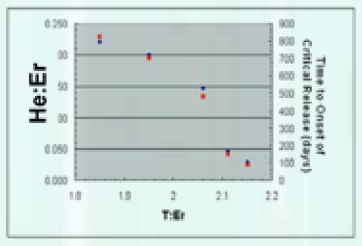
- Hydrogen occluders of the rare-earth and transition metals have complex phase diagrams. The diagram below shows an Er-H isotherm for an erbium powder.
- Three distinct crystallographic phases are possible.





- Helium produced by the radioactive decay of tritium needs to be retained in the target. RFi = Release Fraction

- A link between helium release and target film stoichiometry has been reported.



(From Beavis et al., J. Vac Sci. Technol., **14** (1977), 509)

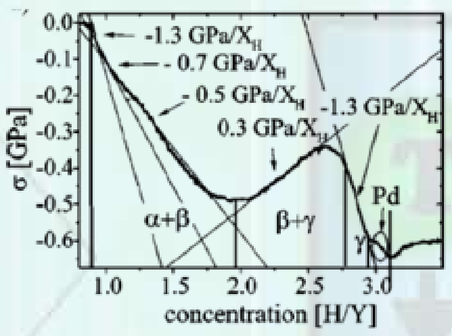
How do you measure the important parameters of metal hydride films?

Use optics to in-situ monitor the hydriding process to determine when the film is

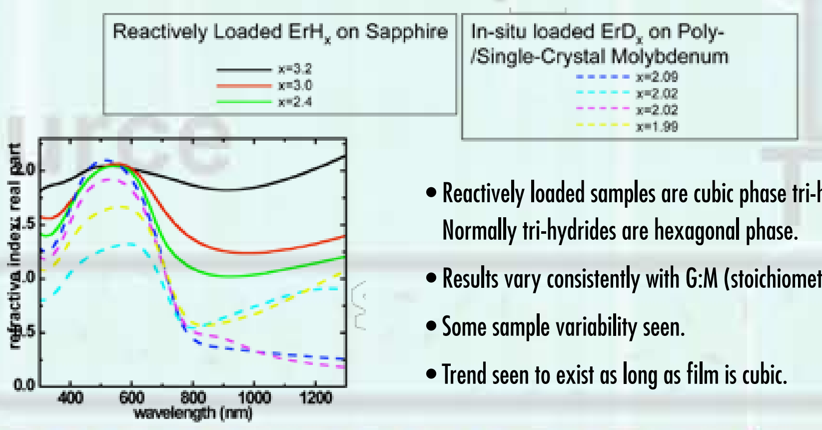
1. in the property crystallographic phase
2. has the right stoichiometry.

- Optical properties of metal hydrides change with film stoichiometry.
- Notice the color change of the target films below for different Gas:Metal ratios.
- Stress state of metal hydride films change with film stoichiometry.
- Example below is for YHx.

G:M = 0 Er metal | G:M = 2.14 β phase | G:M = 2.98 γ phase

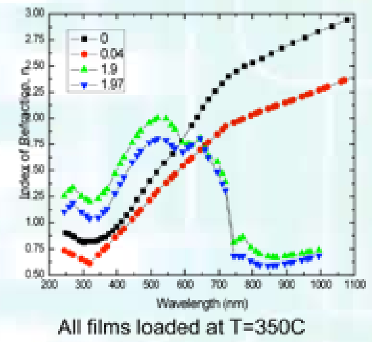


Ellipsometry on smooth reactively loaded ErD_x films



- Reactively loaded samples are cubic phase tri-hydrides. Normally tri-hydrides are hexagonal phase.
- Results vary consistently with G:M (stoichiometry).
- Some sample variability seen.
- Trend seen to exist as long as film is cubic.

Ellipsometry of smooth ErD_x films on Moly/Silicon substrates



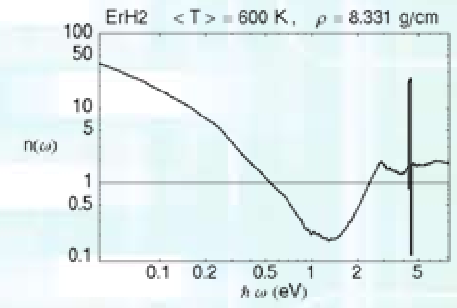
All films loaded at T=350C

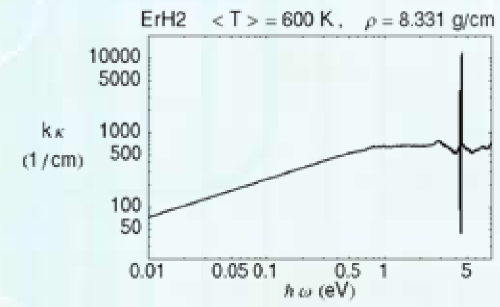
Pressure (Torr)	Measured G:M	Expected G:M
0	0	0
7	0.04	1.85
116	1.9	2.05
336	1.97	2.15

- Same trends seen as from other sample sets.
- Expected G:M predicted from older thermodynamics on powders does not match measured values.
- We have not been able to consistently produce predicted lower stoichiometry films.
  - Are the thermodynamics wrong for films?
  - Films more pure than older films?
  - Differences between powders and films?
  - Are the kinetics driving the problem?

The optical properties of films can also be modeled

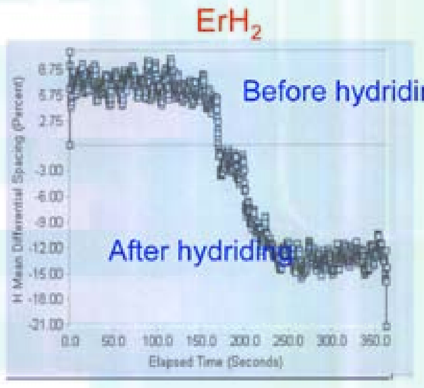
- Technique developed at Sandia by T. Mattsson and M. Desjarlais
- Equilibrium positions of atoms in large 3x3x3 cell computed with molecular dynamics.
- Electronic transitions of large cell computed with very high accuracy using VASP, an ab-initio electronic structure code.
- Optical properties calculated from the electronic transitions using the Kubo-Greenwood formulism.
- Results compare favorable to experimental results in spite of the differences in temperature.

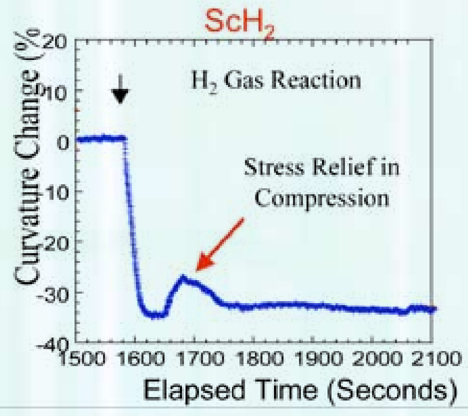




Stress state of film is measured using Sandia developed MOSS

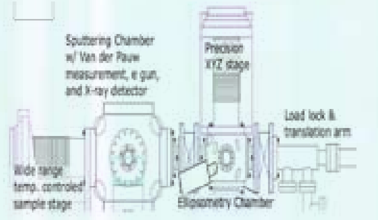
- MOSS=Multi-beam Optical Stress Sensor
- Measures curvature of substrate
- Thin film after deposition is in tensile stress
- Upon heating/hydriding film approaches zero stress
- After hydriding and cooling film develops in-plane compressive stress

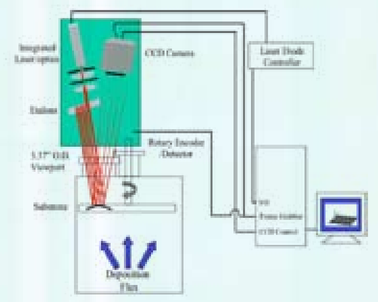




Significance and impact of results

- Our results call into question the validity of the thermodynamics/kinetics used to produce neutron tube targets.
- We are using the optical techniques developed in this LDRD to answer this critical question.
  - We are performing in-situ spectroscopic ellipsometry and reflectivity to monitor the uptake and release of hydrogen at differing temperatures and pressures.
  - We are performing MOSS measurements as film is hydriding to monitor the uptake and release of hydrogen at differing temperatures and pressures.





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