Science-led Materials Discovery

High Throughput Discovery of Novel Complex Metal Hydrides

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SYNTHESIS

Methodology

High Throughput Physical Vapour Deposition Systems

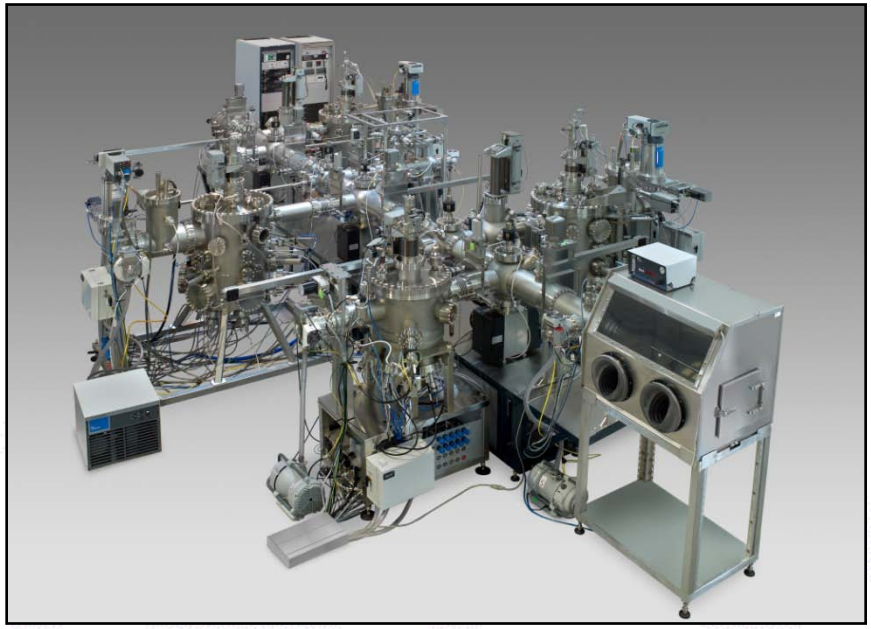

Image DCA ©

UoS PVD

KDH PVD

Generic Schematic of synthetic systems

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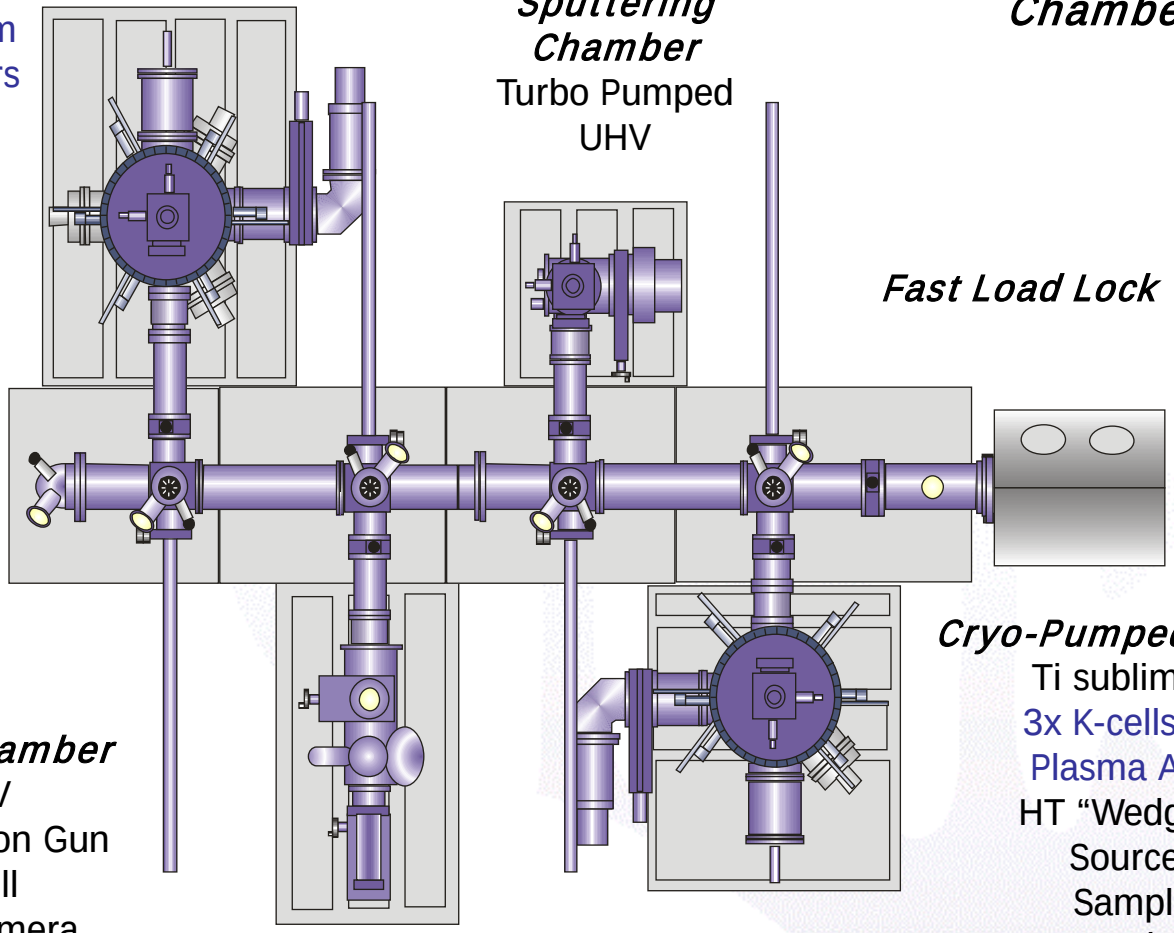
UHV System with Sample Travel under UHV Between Chambers

Cryo-Pumped MBE Chamber
Ti sublimation Pump
3x K-cells, 3x e-beam
HT "Wedge " Shutters
Source Shutters
Sample Shutter
Heated Manipulator

Sputtering Chamber
Turbo Pumped
UHV

Fast Load Lock

Masking
Station



Glove Box

Cryo-Pumped MBE Chamber
Ti sublimation Pump
3x K-cells, 1x e-beam
Plasma Atom Source
HT "Wedge " Shutters
Source Shutters
Sample Shutter
Heated Manipulator

Surface Analysis Chamber
Ion Pumped UHV
Spot-XPS, AES, TPD, Ion Gun
High Pressure Cell
Thermal Imaging Camera

Guerin and Hayden; J. Combinatorial Chemistry 8 (2006) 66-73

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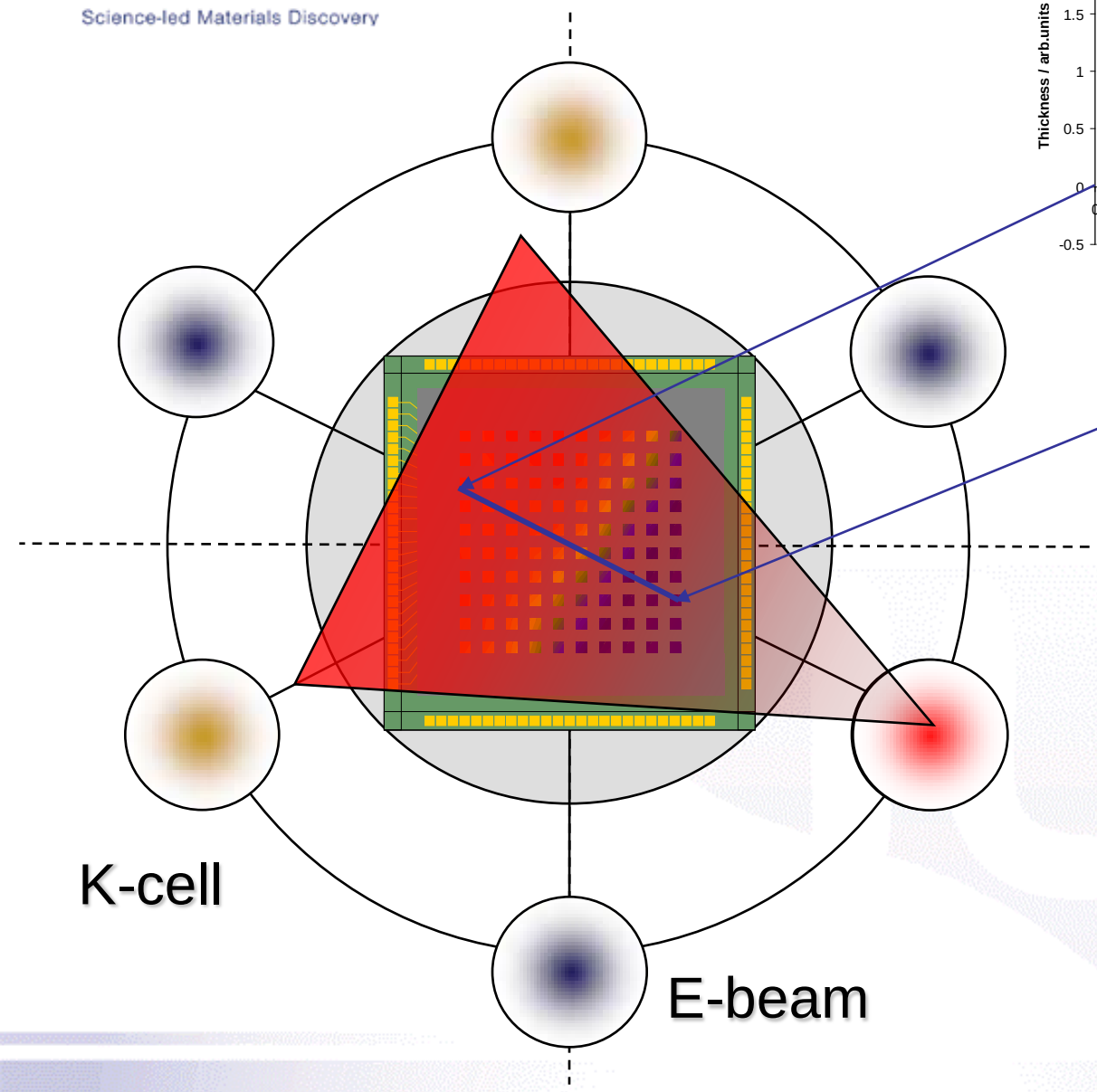

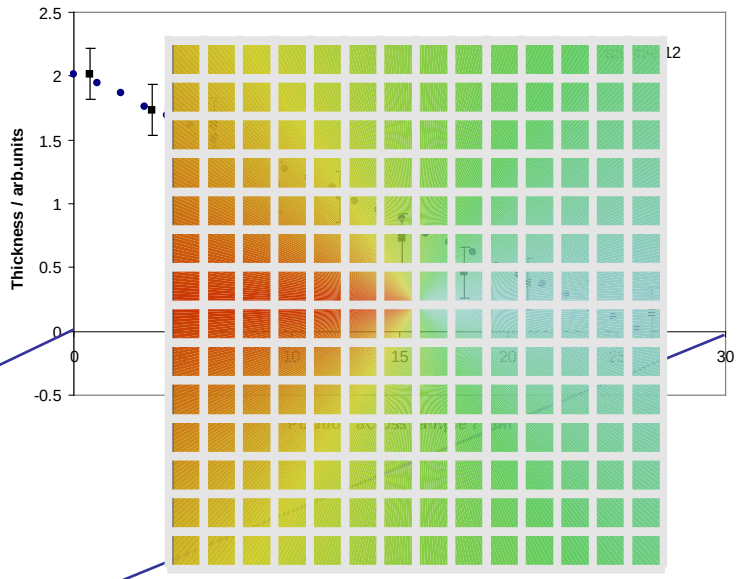

Simultaneous Deposition of Elements from Multiple Sources with control of the Contact Masks

The "Wedge" Profile and Rates results in Thin Film Materials with Compositional Gradients

Thin Film Materials are confined to "fields" on screening chips using Contact Masks

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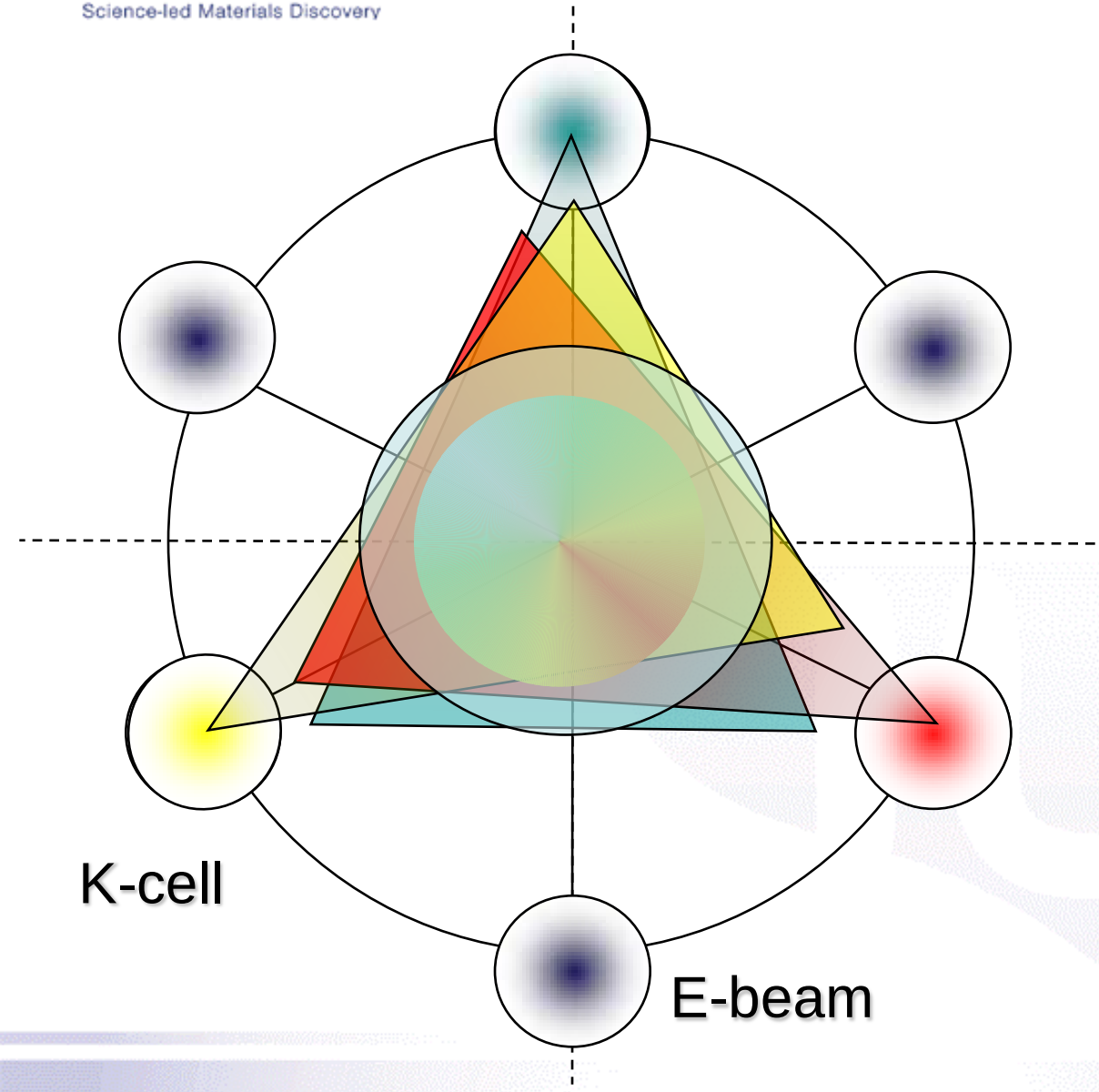

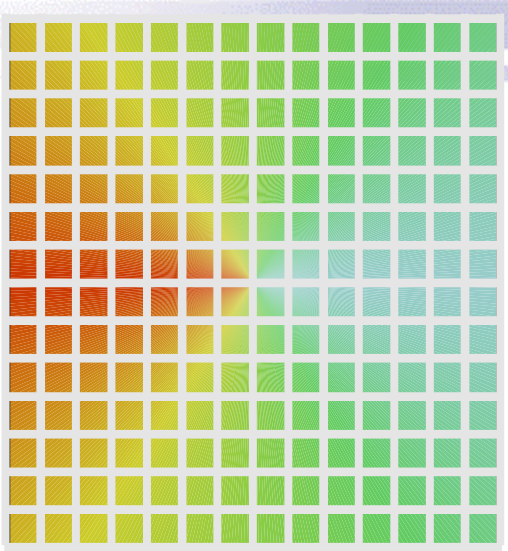

In-Situ Plasma Source Provides Atomic Hydrogen, Multiple Sources, with Hydrogen to "Produce" Hydride, Nitride or Oxide Phases

Deposition of Elemental Oxygen, Hydrogen to Produce Hydride, Nitride or Oxide Phases

Ratio Results in Thin Film Materials with Compositional Gradients

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HT-PVD Synthesis

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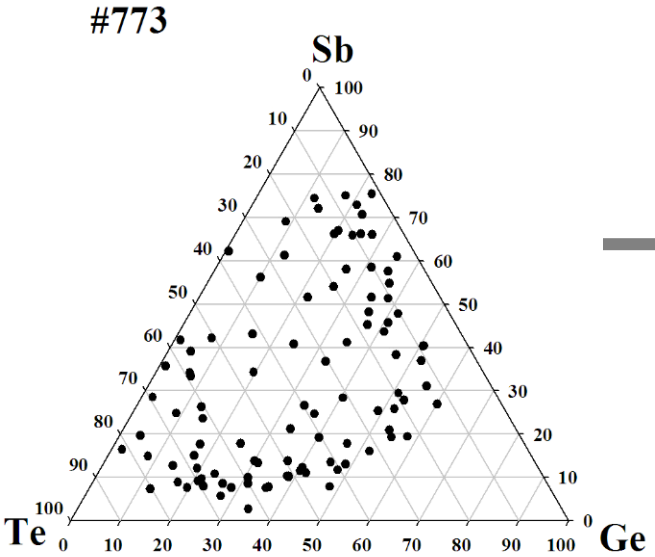
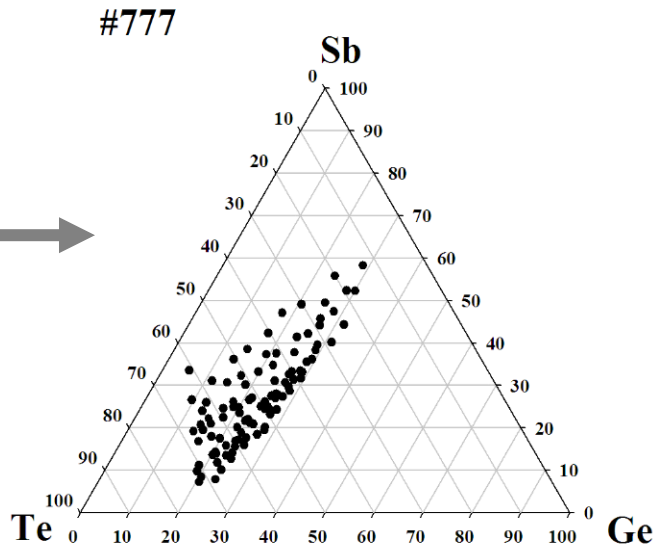
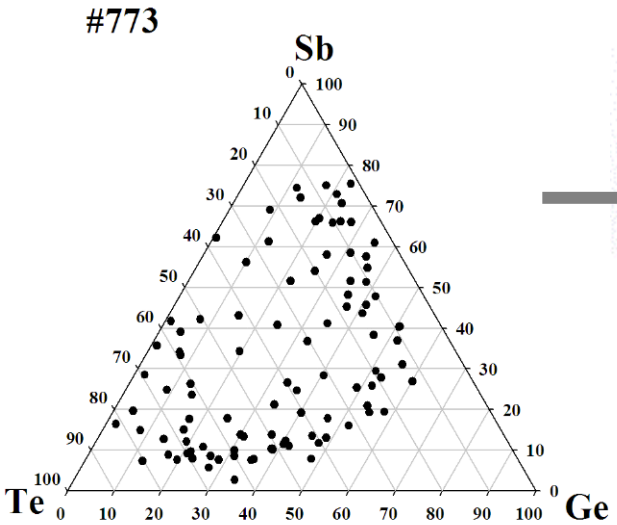
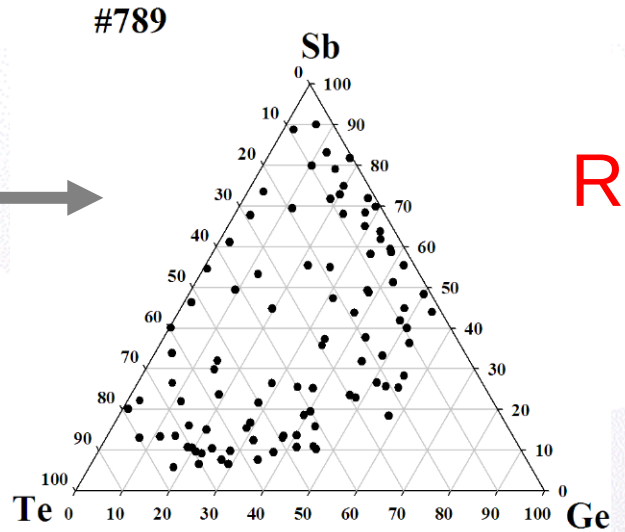
Control









Reproducibility

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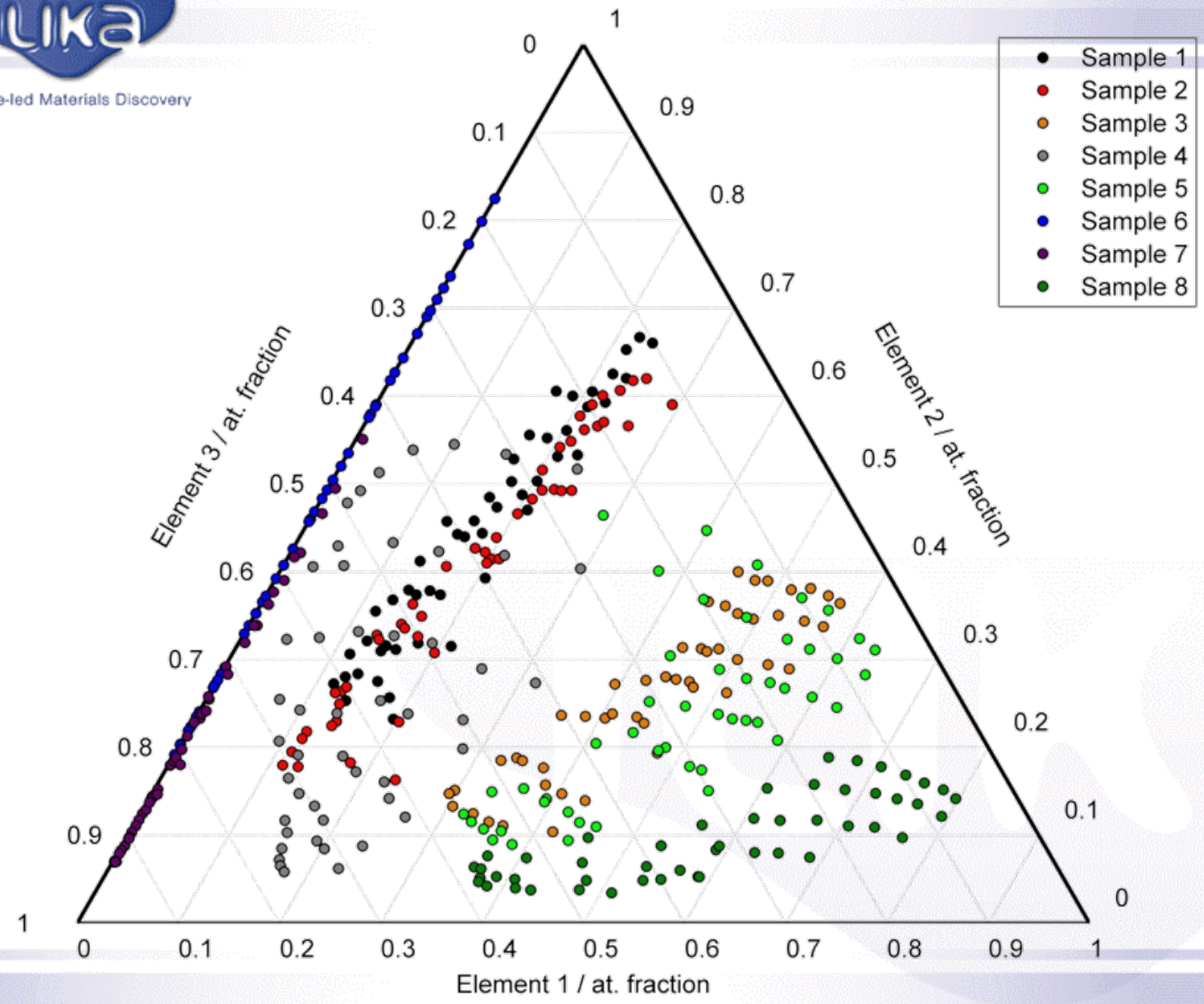

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CHARACTERISATION AND SCREENING

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Parameter	Equipment
Composition And Structure	Automated SEM and EDS Automated Thin Film X-ray Diffraction Automated Atomic Force Microscopy High Resolution TEM and EDS Automated Raman Microscopy Automated Infrared Microscopy Automated Optical Profilometer Automated Spot X-Ray Photoelectron Spectroscopy Automated Laser Ablation Inductively Coupled Mass spectroscopy
Conductivity	Automated Four-Point Probe or Van de Pauw
Optical	Automated Imaging Ellipsometer Imaging Reflectivity Automated Visible/UV Transmission Spectroscopy
Thermal	Imaging Thermal (IR) Camera
Thickness	Optical Profilometer Automated Atomic Force Microscopy Automated Imaging Ellipsometer
Hydrogen storage	Automated Pressure – Temperature - Transmission measurements High throughput Temperature Programmed desorption

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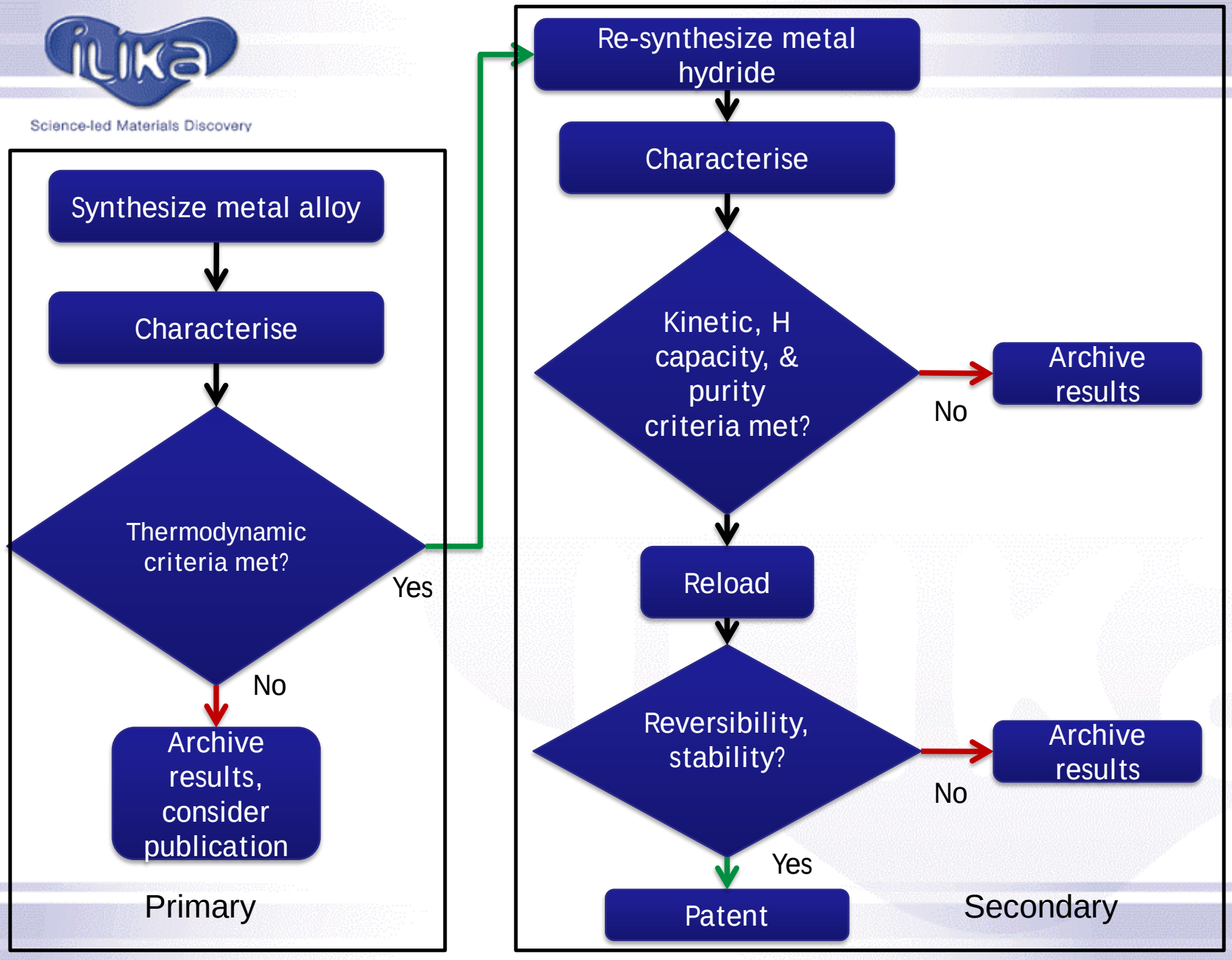
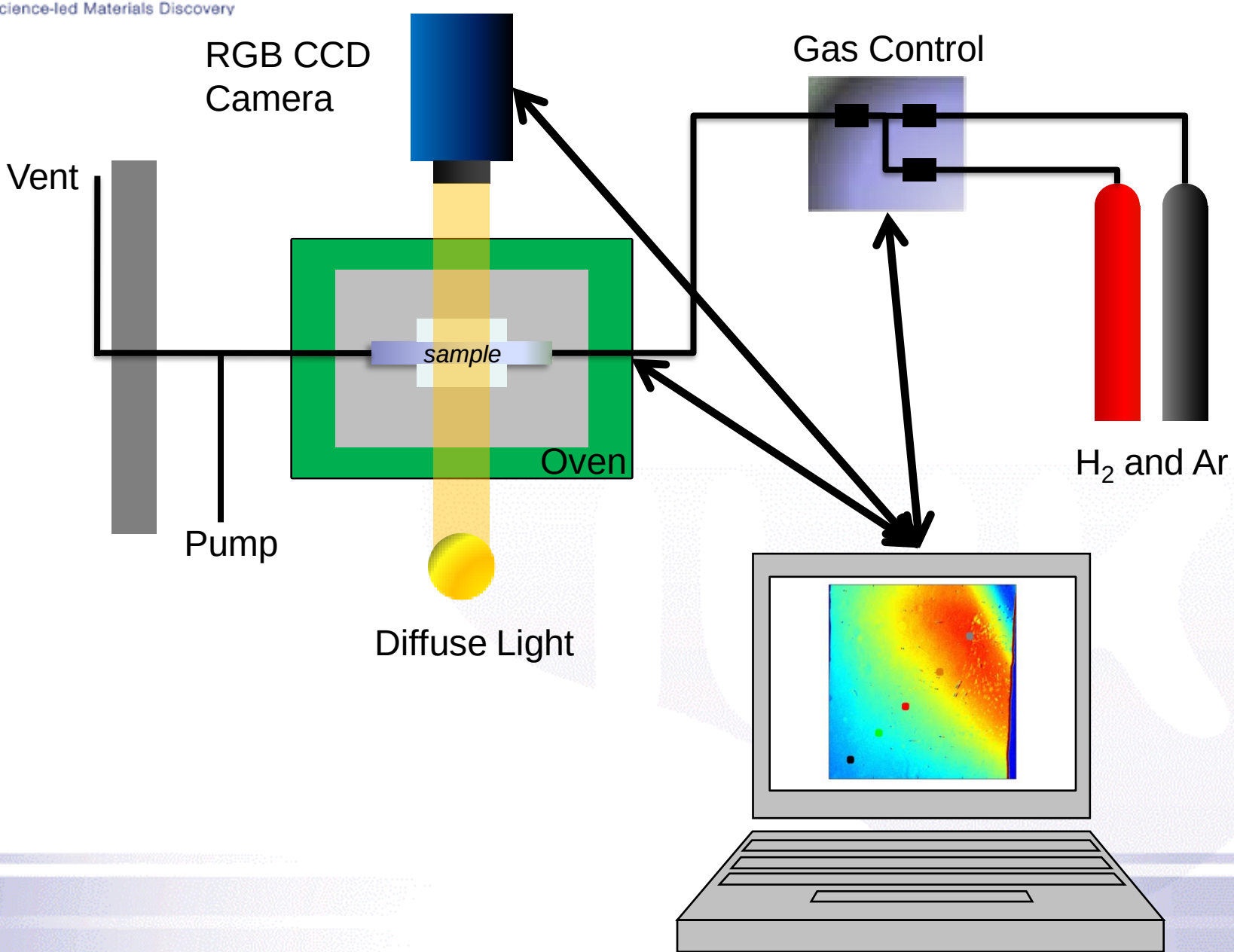


Summary of primary screening: P-T-T

Pressure-Temperature-Transmission



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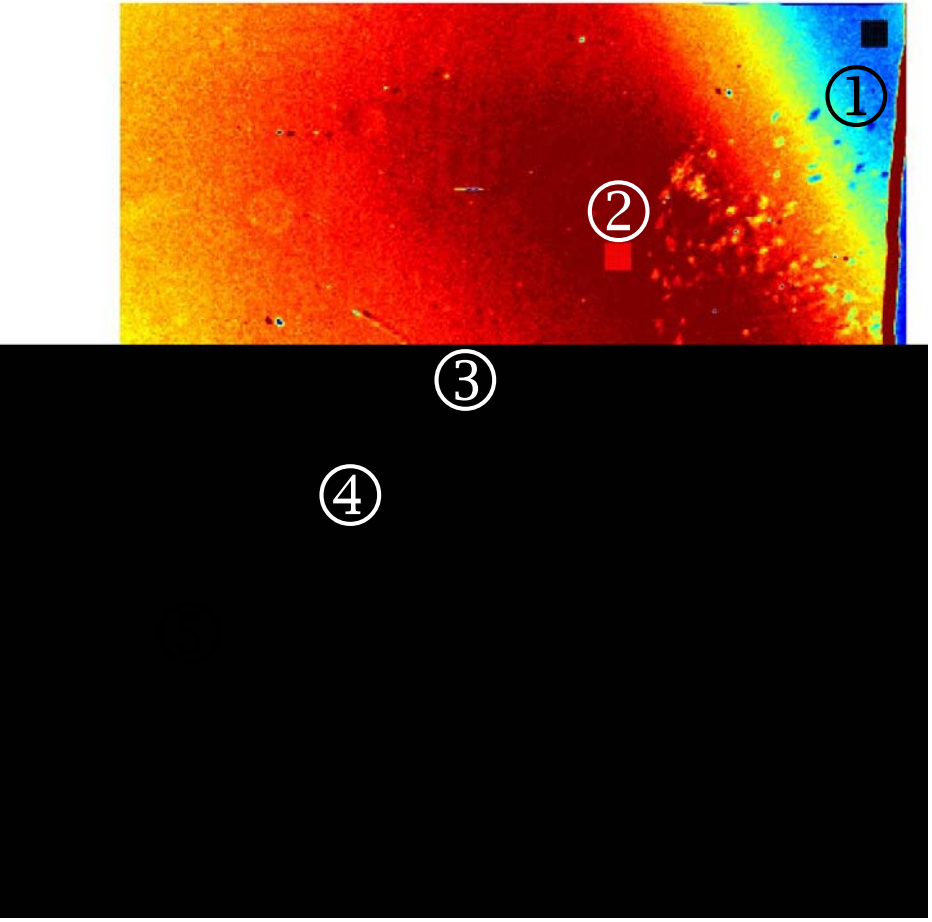
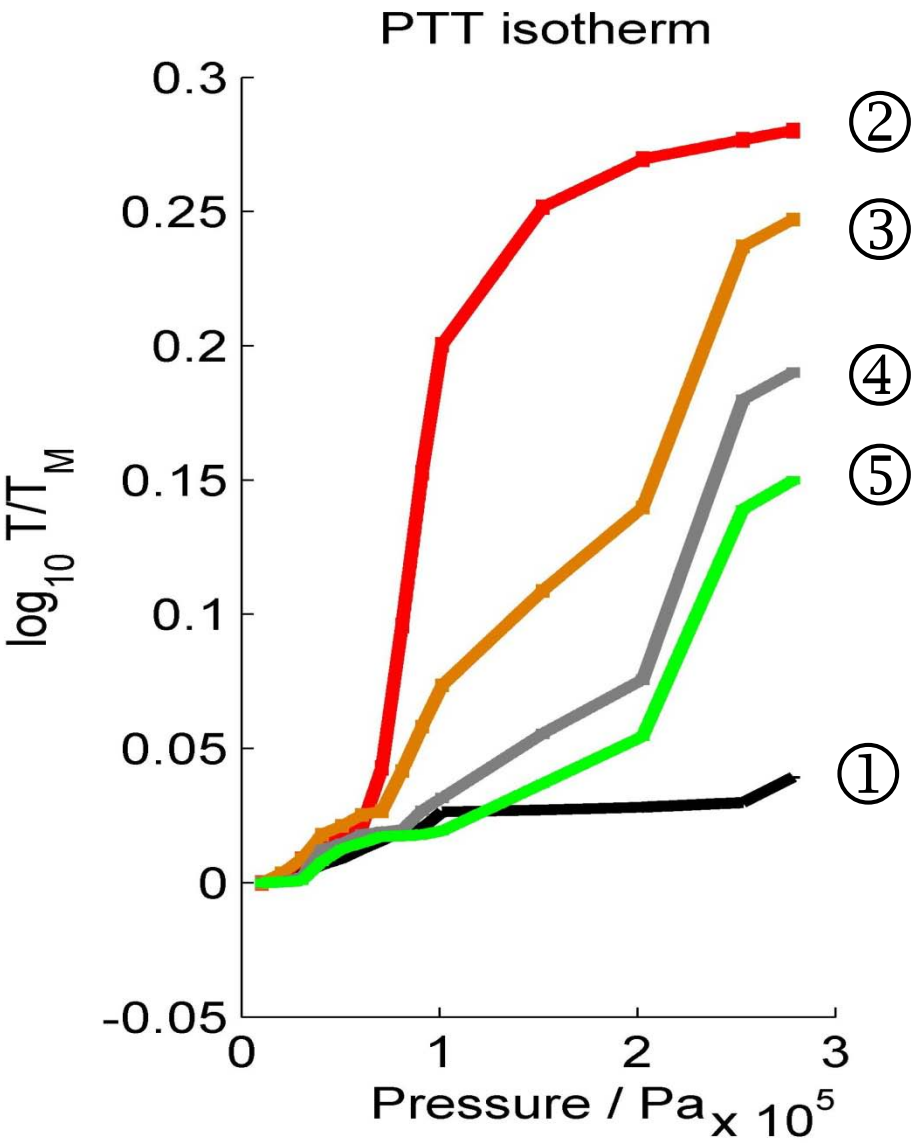
Δ Transmission





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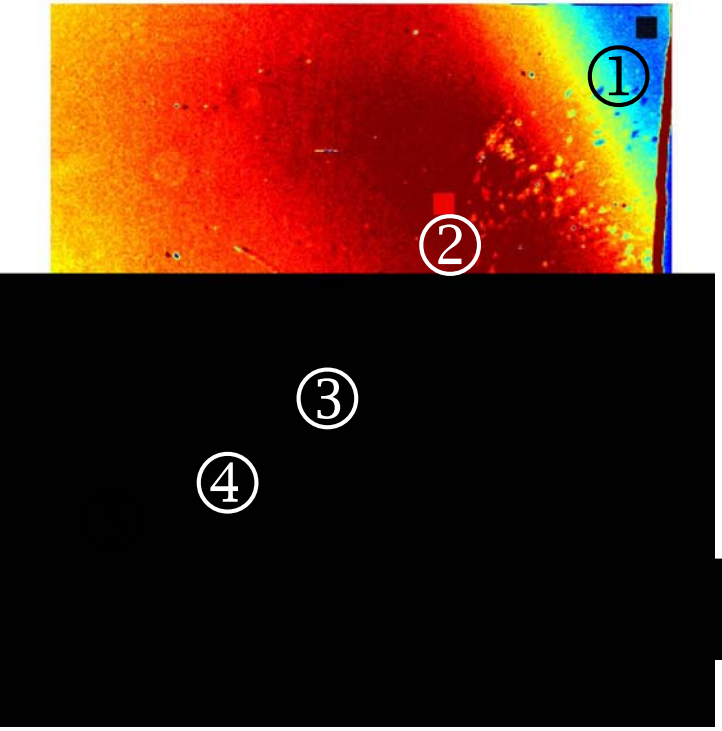
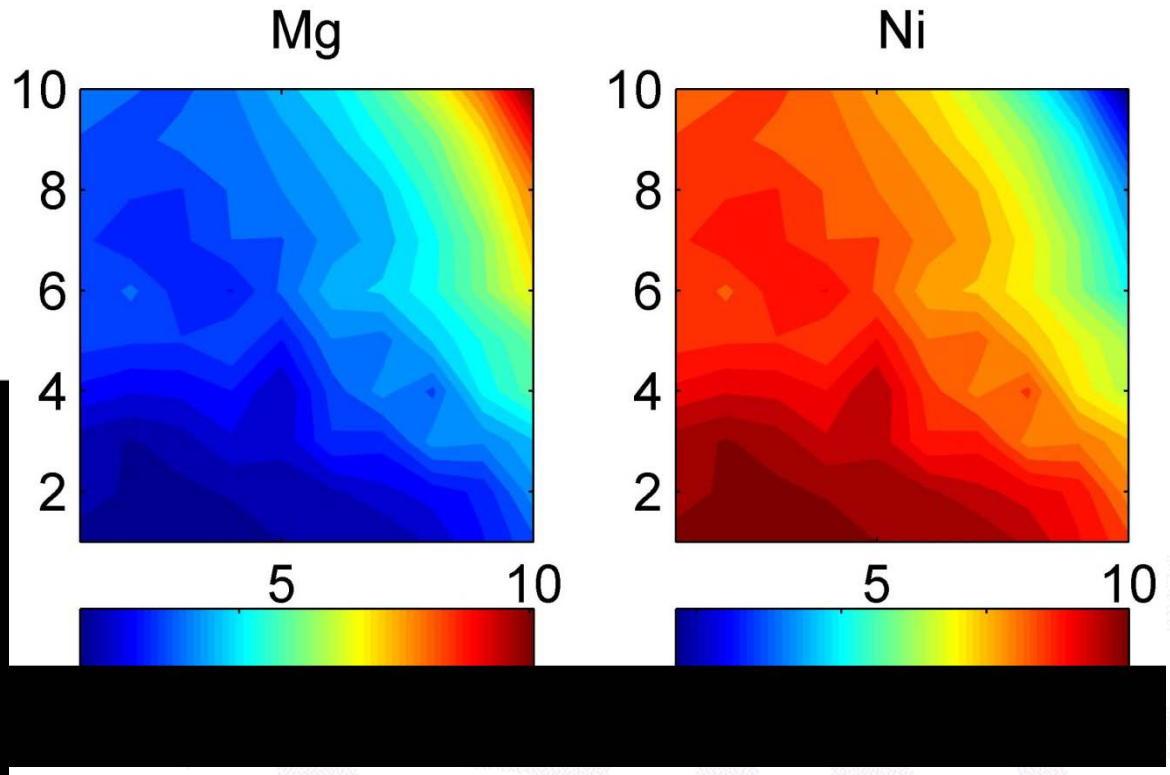
Δ Transmission





Red Spot (②) is $Mg_{0.69}Ni_{0.31}$

Plateau pressure = 0.5 – 1.0 x 10^5 Pa at 175 ^oC

Mg-Ni Hydrogenation Cycle

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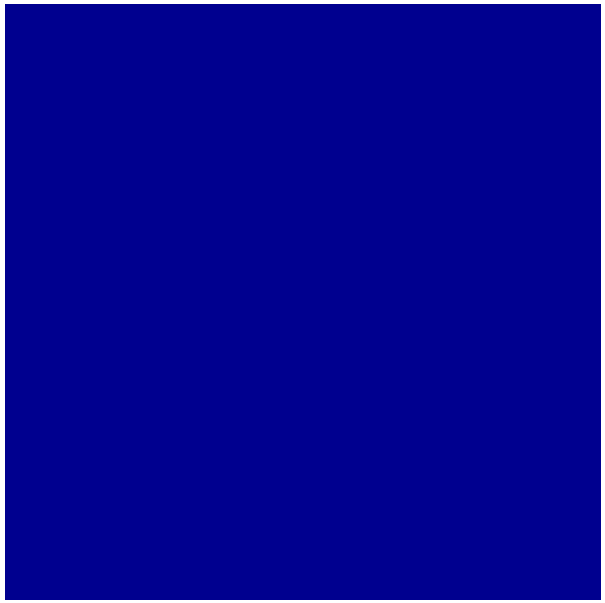
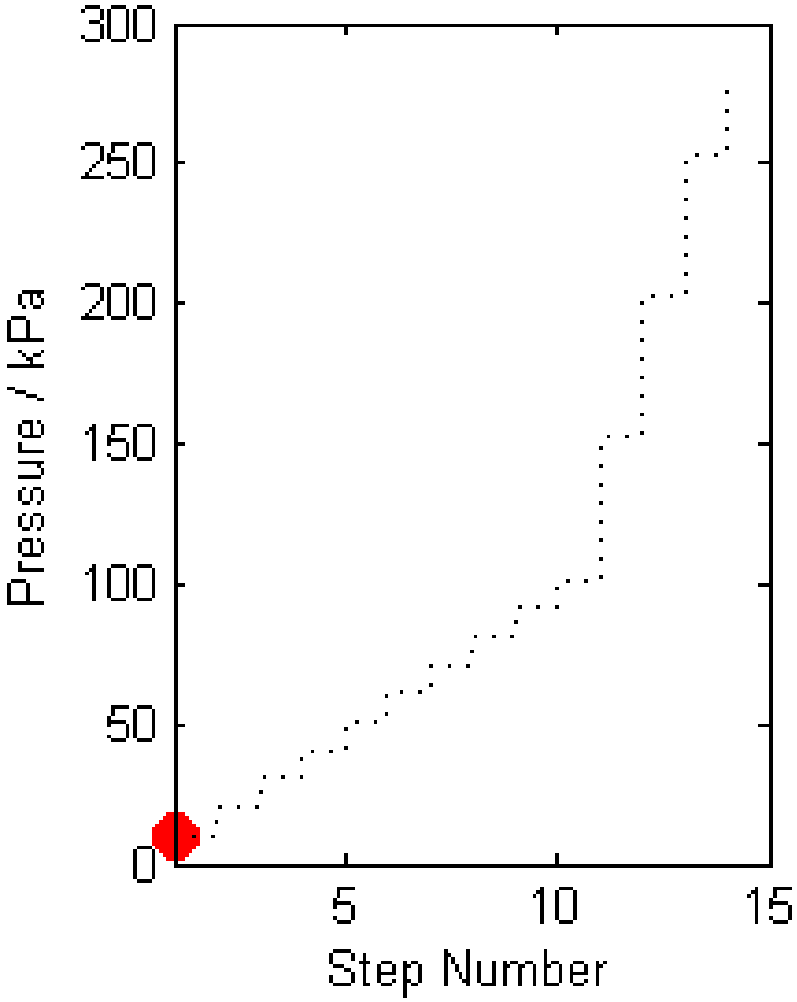
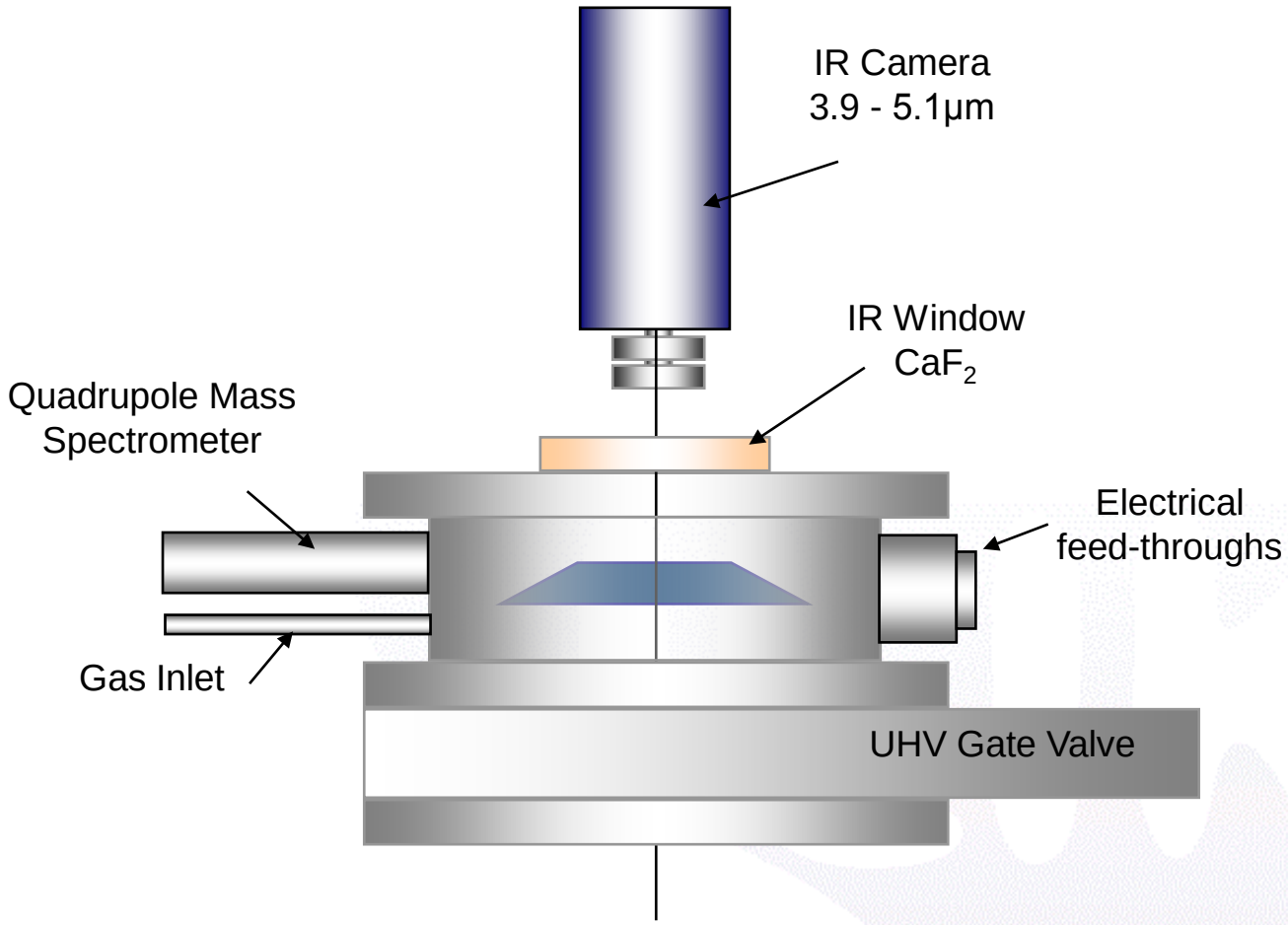
Pressure : 11 kPa





Summary of secondary screening: TPD

Temperature Programmed Desorption & Reversibility



Summary of secondary screening: TPD

Temperature Programmed Desorption

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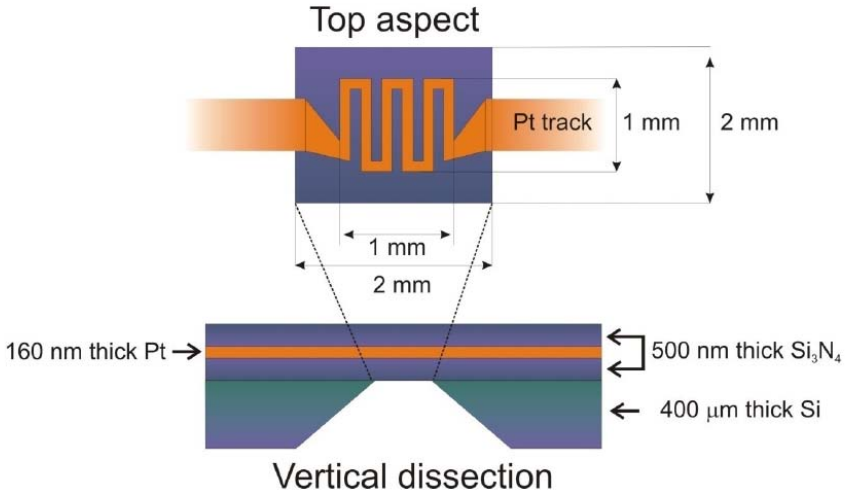
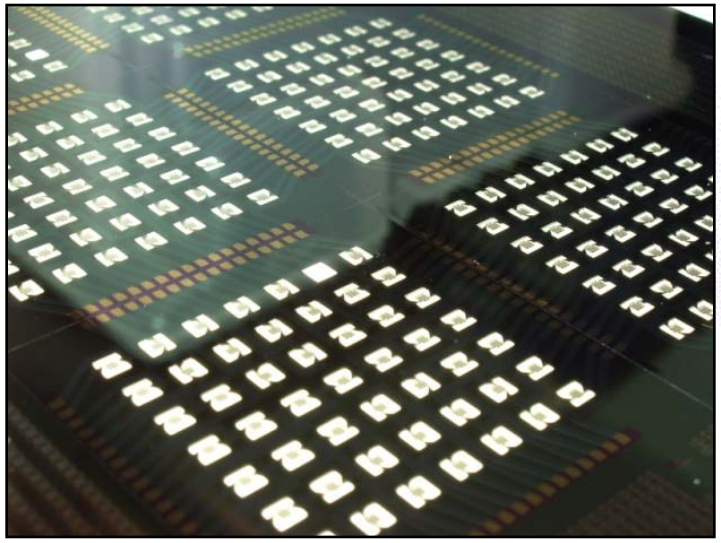
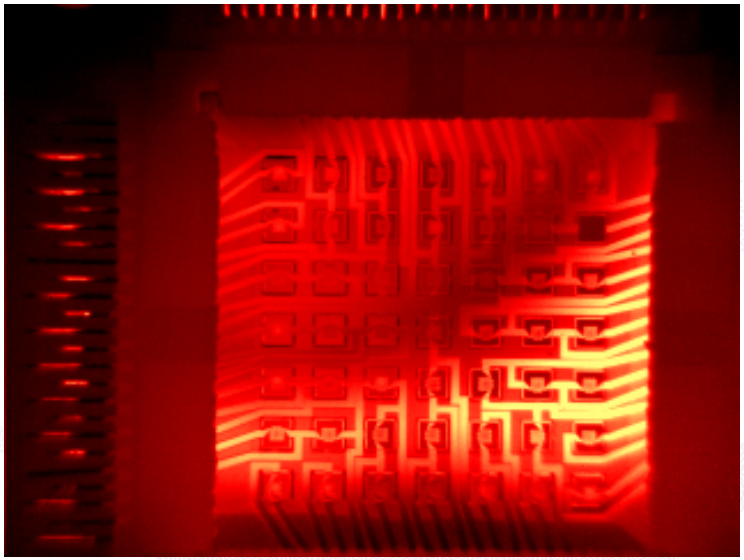
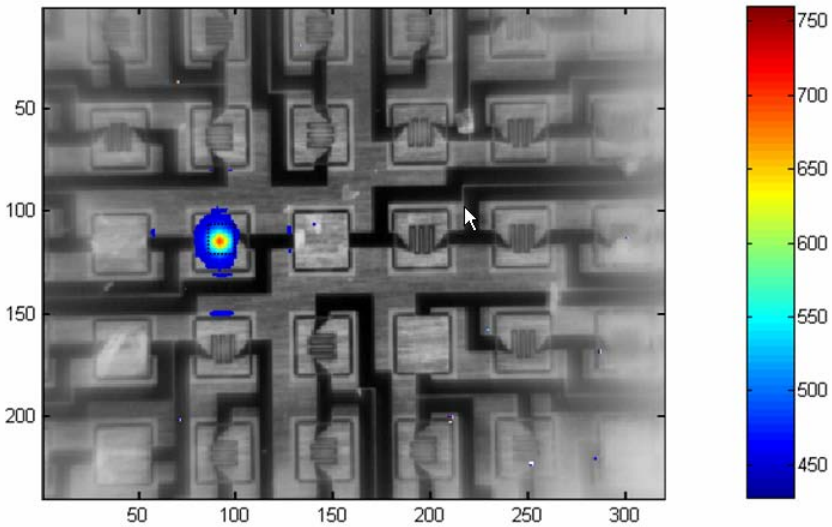
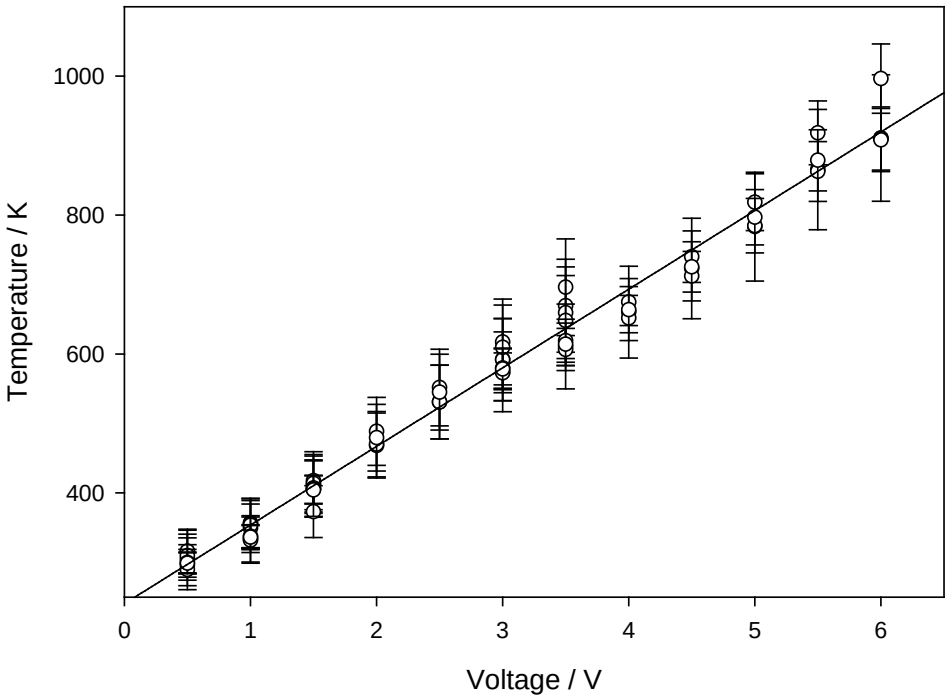






MEMS Array





Vacuum heating rate determined to be **114** 6 K V^{-1}

Maximum temperature therefore **957** K 36 K V^{-1}

TPD: Magnesium – Nickel Hydrides

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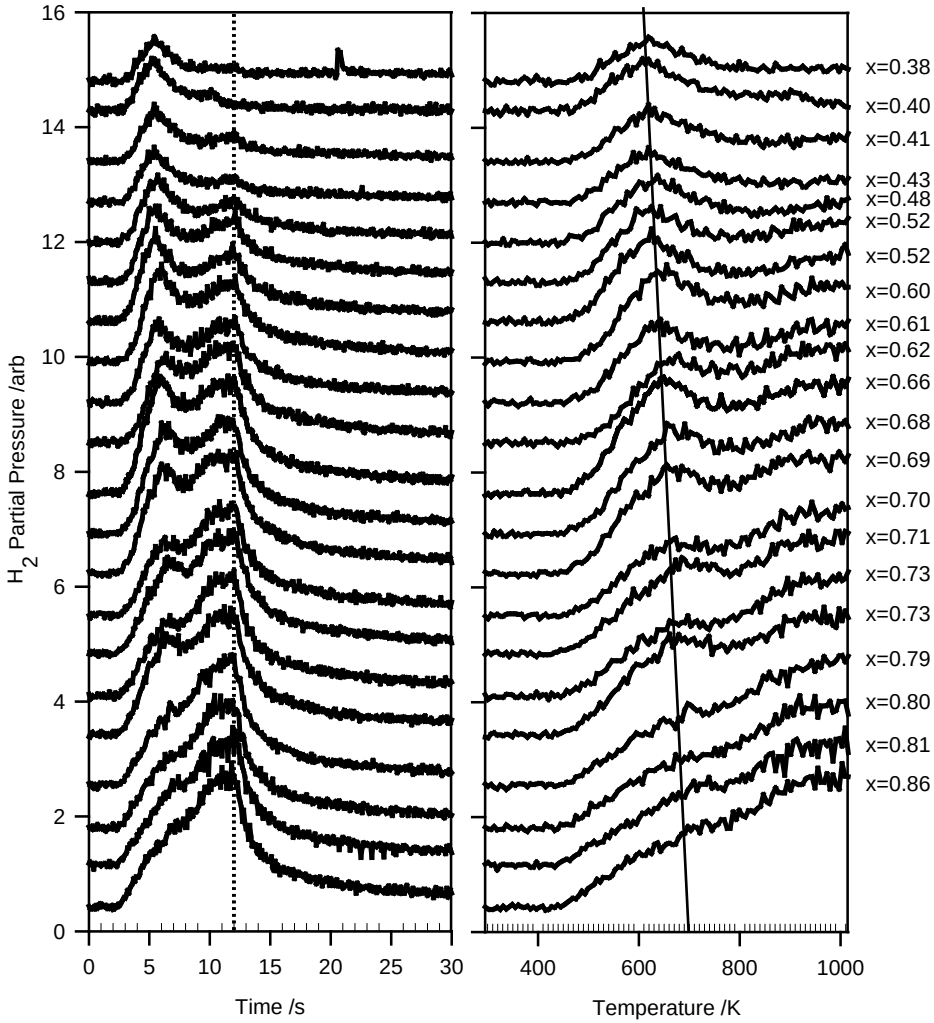
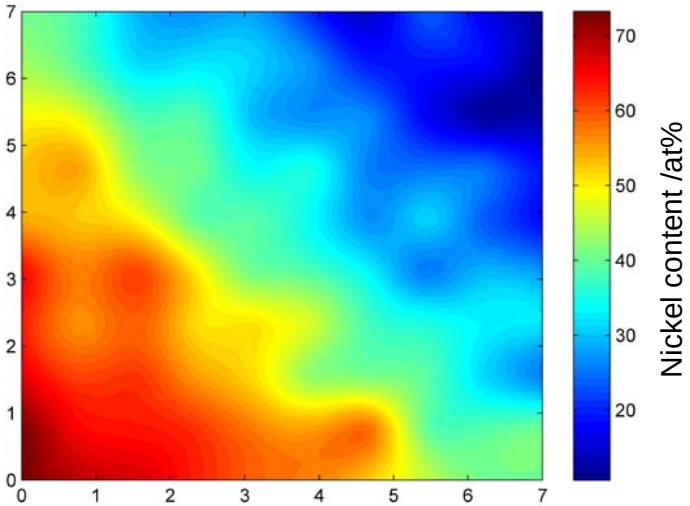
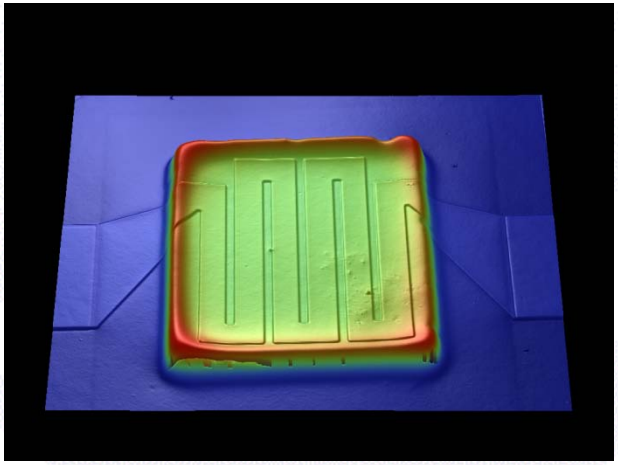
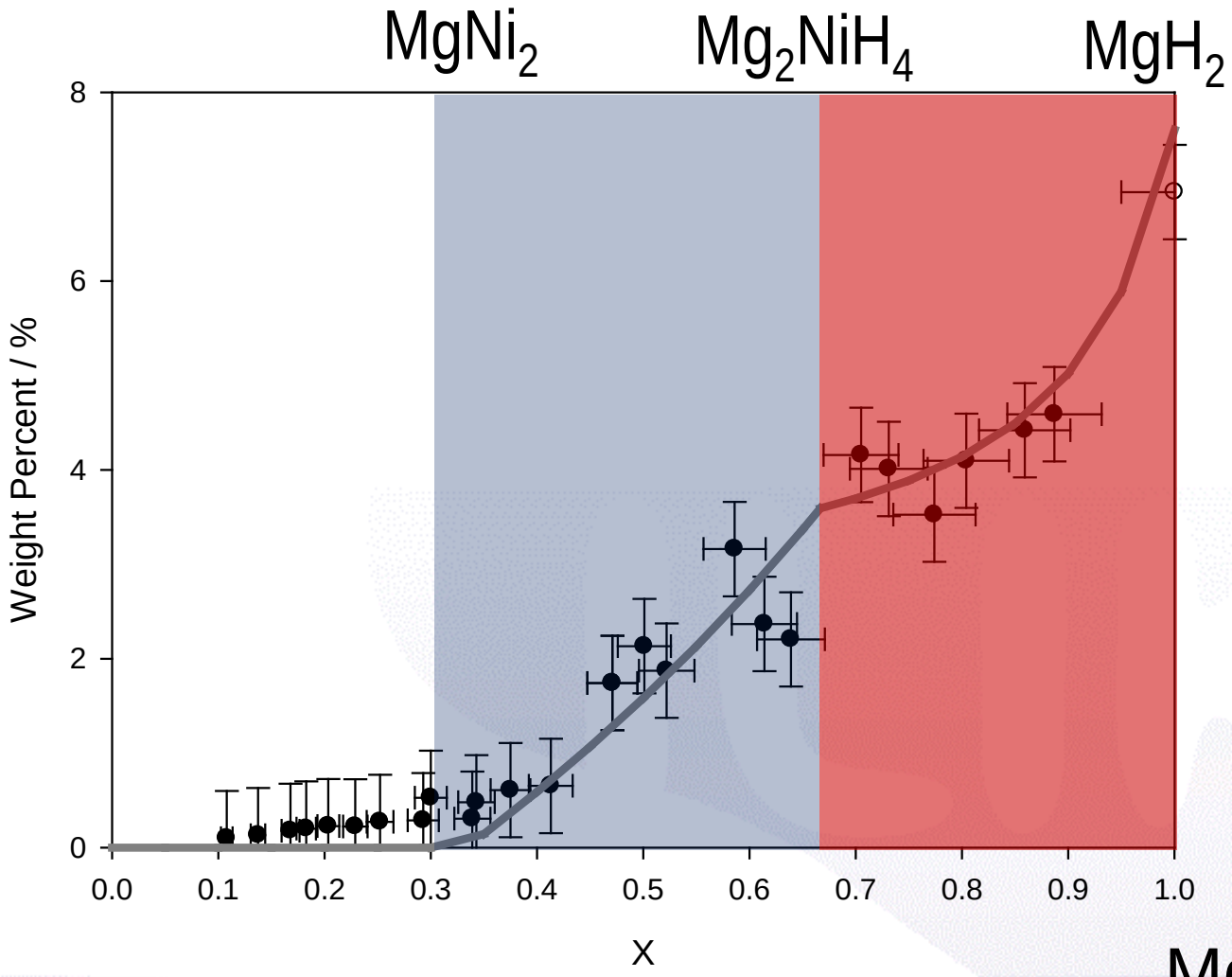




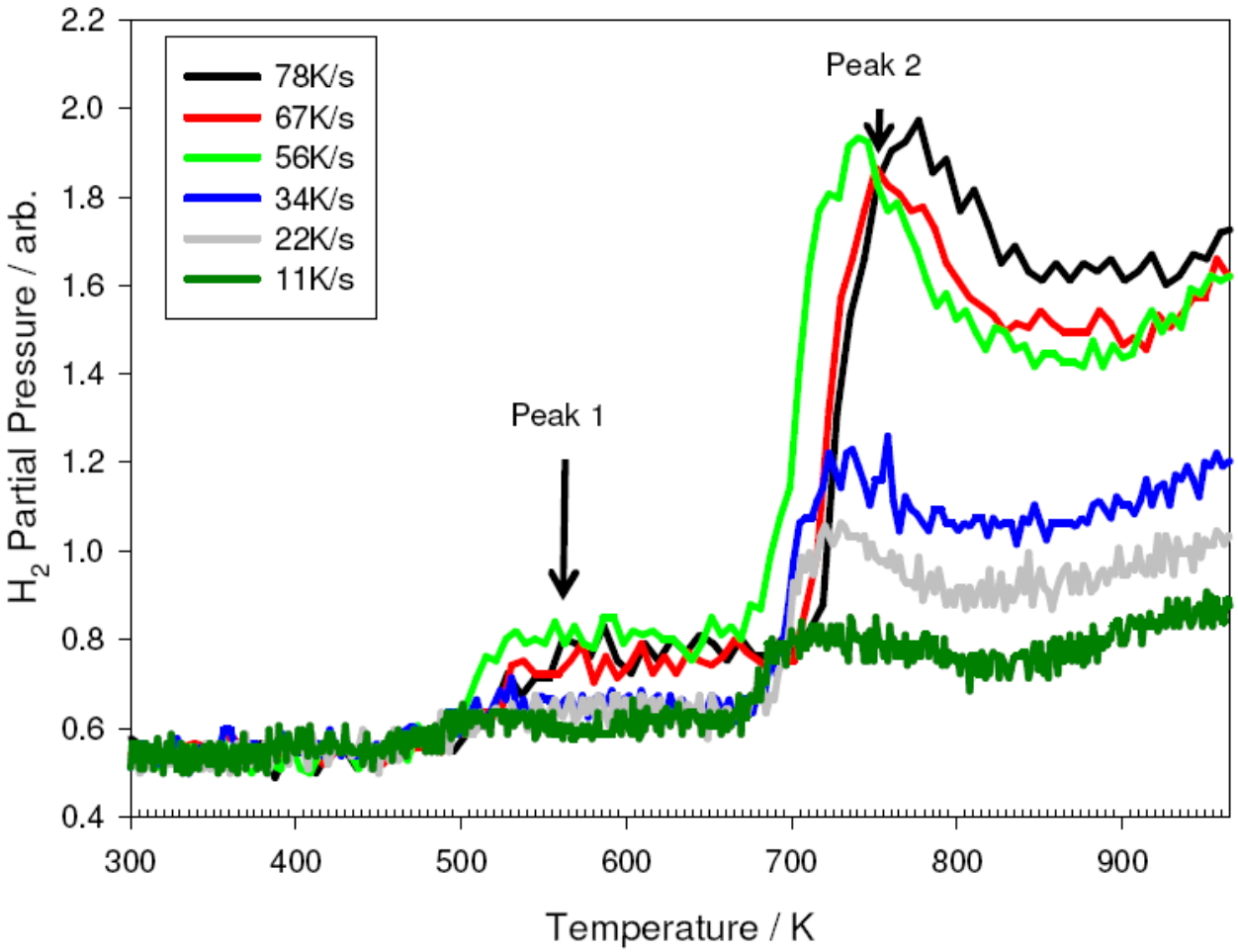


Magnesium – Nickel Hydrides



$Mg_xNi_{1-x}H_y$

TPD



$Mg_{0.8}Ni_{0.2}H_y$

Apparent Activation Energies

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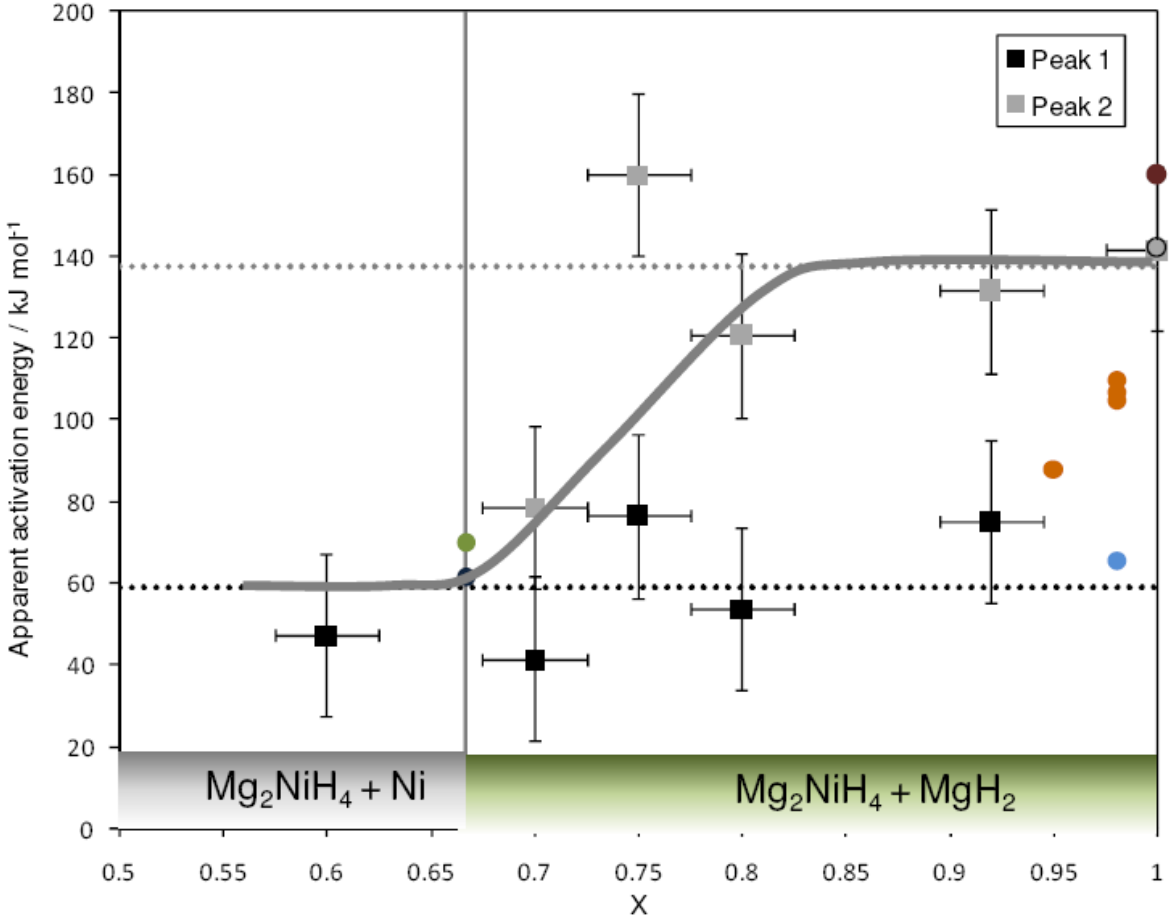

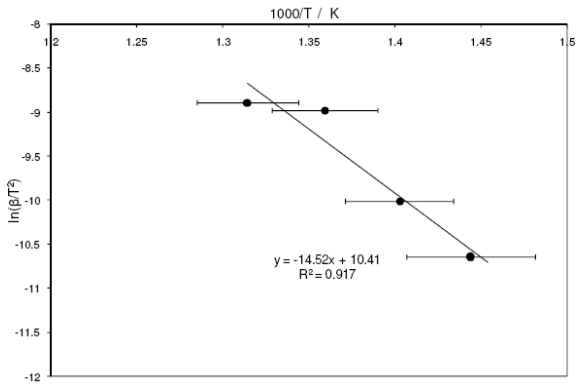

$Mg_{0.8}Ni_{0.2}H_y$

Summary of Figures of Merit

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All FOM determined as a function of composition

- Primary Screen:
  - ΔH, ΔS
- Secondary Screen:
  - Gravimetric capacity (wt. %)
    - Initial and reversible
  - T_{onset} @ given heating rate
  - E_a
  - Stability (over *n* cycles)

Complete picture of hydrogen sorption for lead candidate identification

Summary

- Synthesis
  - Patented approach to high throughput synthesis of materials
- Screening
  - PTT and TPD combine to yield a complete picture of hydrogen sorption and storage
- Informatics
  - High throughput analysis provides means to rapidly visualise and interpret results