Ceramic Scintillator Alpha Detector for Advanced D-T Generator Neutron Tagging

OR07-AI-AlphaD-PD03

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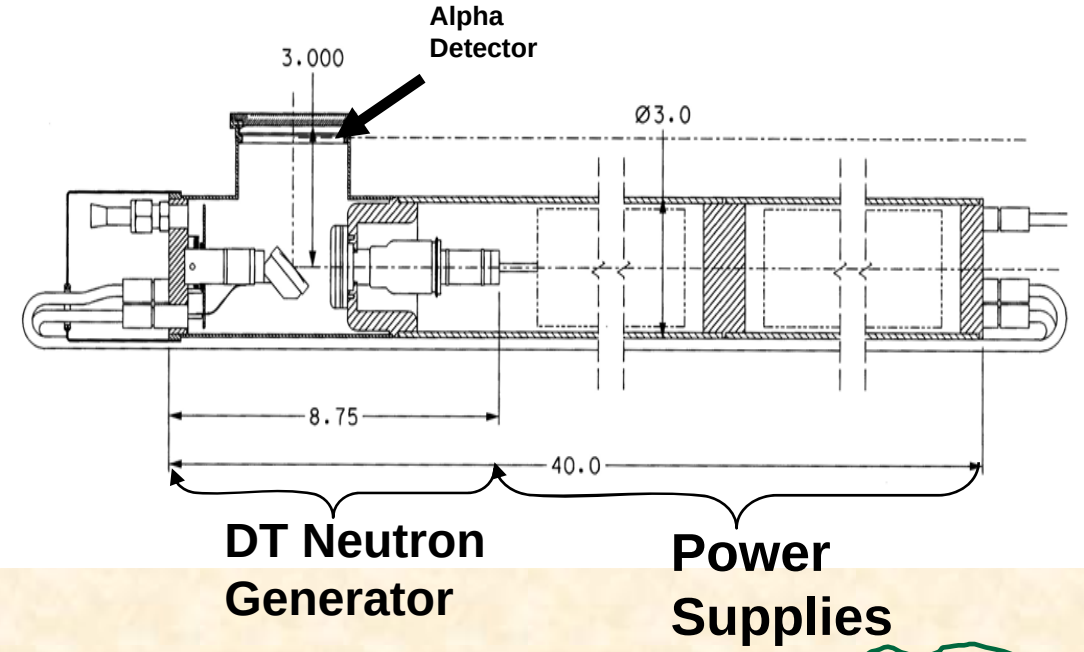
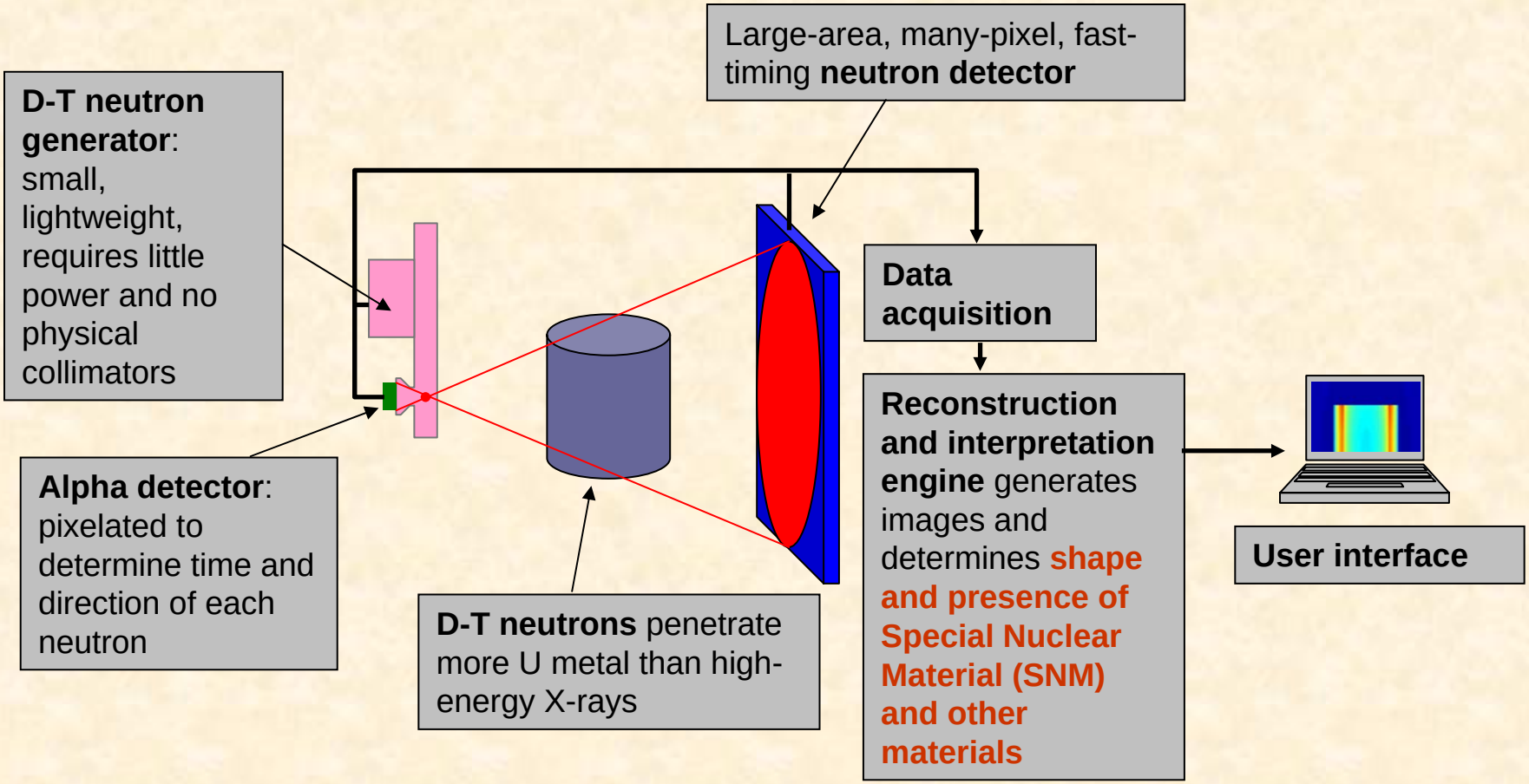
Outline of Talk

- **Project description**
- **State of the art**
- **Project plan and accomplishments to date**
- **Summary**

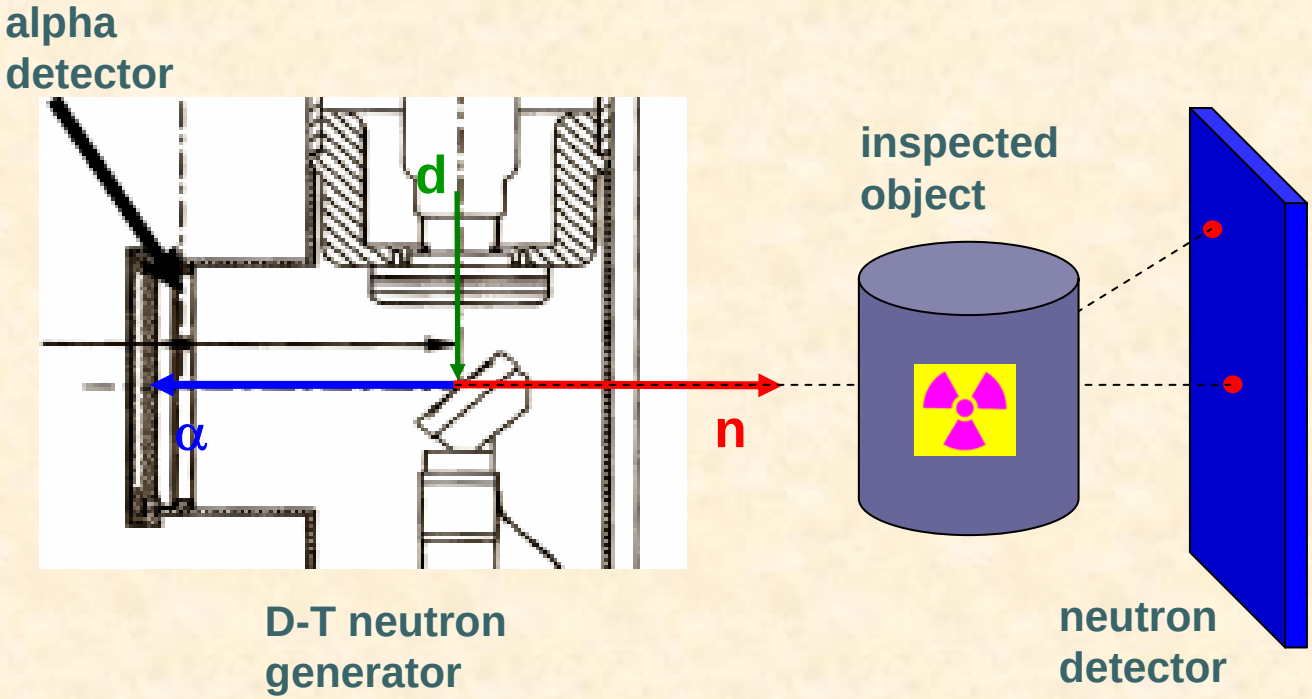


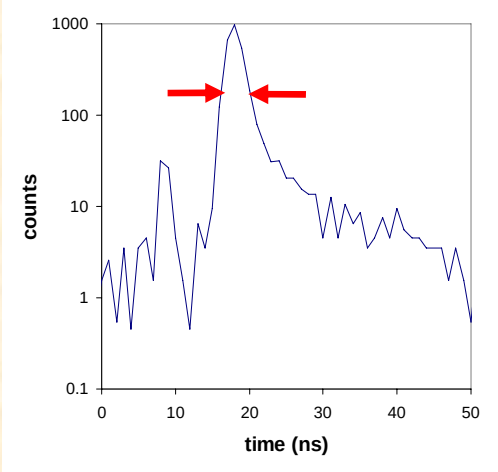
Fast Neutron Imaging System Concept



Arrival of 14 MeV neutrons at known time eliminates physical collimator

Principle of Operation



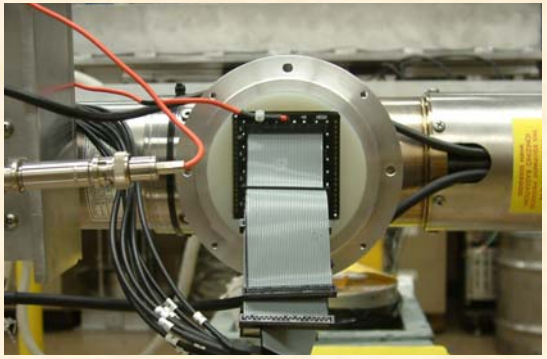


- **Classify counts in neutron detector (transmitted, scattered, or induced neutron, induced gamma) by arrival time and location**
- **Location measured by constructing detectors as a composite of 10,000 neutron-detector pixels and 256 alpha-detector pixels**

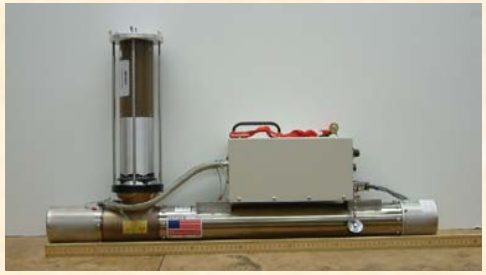
Neutron Imaging System
State of the Art: Evolution of Alpha Detectors

ZnO:Ga Polycrystalline Ceramic
Scintillator Detector, Spherical
Pixelated, Electronic Change

This Project



YAP:Ce Single Crystal
Scintillator Detector,Flat
Pixelated, Electronic Change

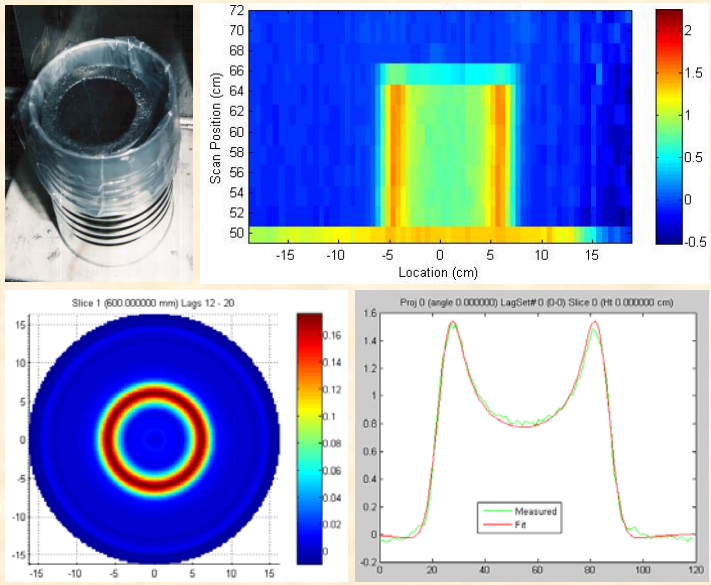


YAP:Ce Single Crystal
Scintillator Detector,Flat
Single Pixel, Manual Change

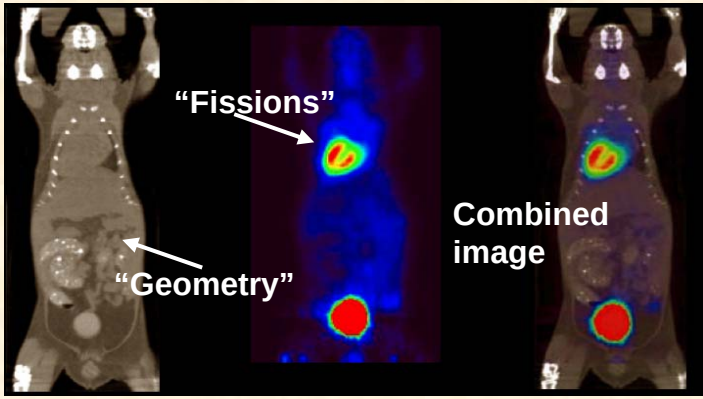
ZnO:Ga Powder
Scintillator Detector, Flat
Single Pixel, Manual Change

Support of ORNL Neutron Imaging System

- **Modifications to the present neutron imaging system could lead to substantial gains in overall system performance and functionality:**
  - **a reduction in the D-T generator beam spot size**
  - **pixelation of the imaging detectors**
  - **pixelation of the associated-particle alpha detector**
  - **improved timing of the associated-particle alpha detector**
- **The improvements in system performance include the following:**
  - **reduced measurement times with comparable position resolution**
  - **better penetration of dense portions of interrogated items**
  - **materials identification**
  - **determination of the functionality of an object (e.g., induced fissions)**
- **This proposal addresses the fabrication of the alpha particle detector using an ultra-fast inorganic scintillator, ZnO:Ga, in a transparent polycrystalline ceramic form. The alpha particle detector would retain the desirable qualities of ZnO:Ga (inorganic and fast) while significantly improving the scintillator light output and enabling uniform, reliable production of alpha detectors.**



Example co-registered CT and PET (Siemens)



Project Plan

ZnO:Ga precursor powder synthesis

- Ga concentration optimization
- Hydrogen treatment optimization

Characterization of precursor powders

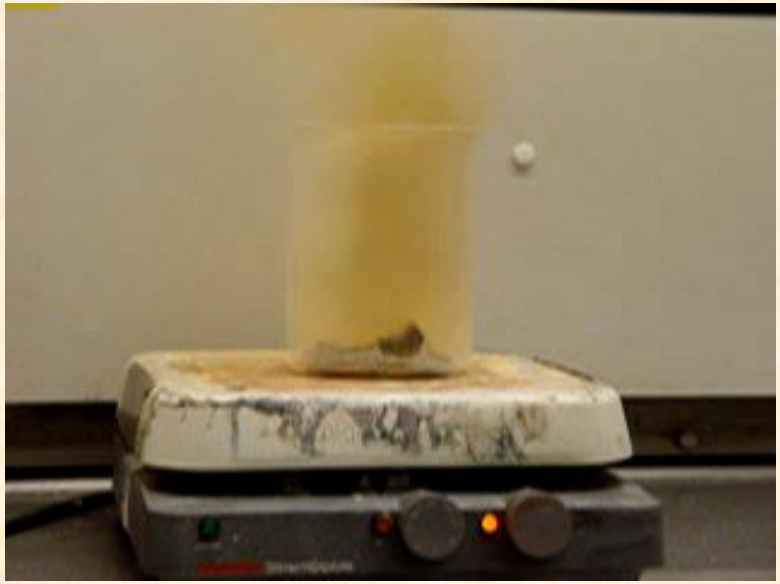
Densification of precursor powders

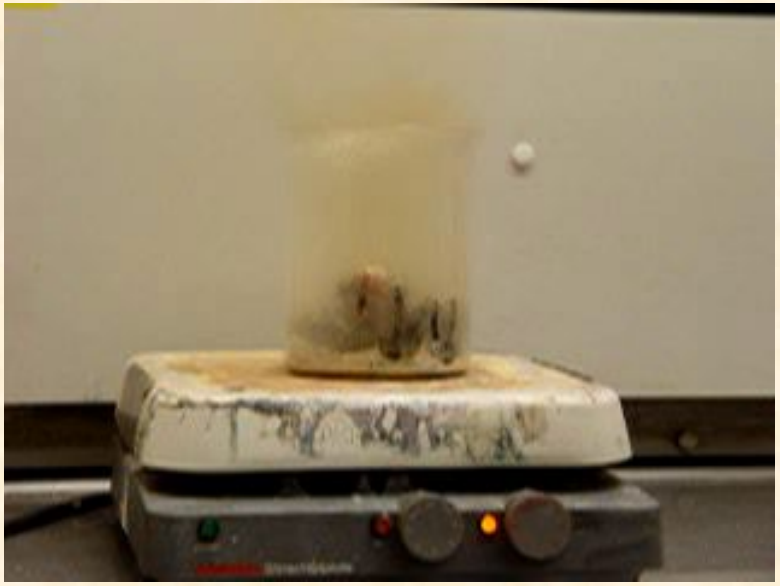
Characterization of densified bodies

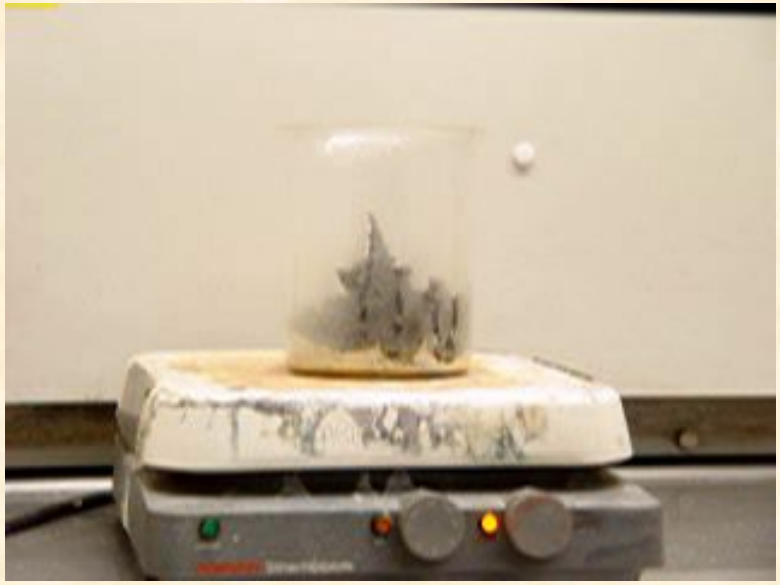
Powder Synthesis

- **Common goals**
  - **Control size, shape, doping**
  - **Chemical purity**
  - **Phase purity**
- **Pursuing three methods of synthesis**
  - **Urea precipitation (ORNL)**
  - **Combustion (ORNL and UC-Davis)**
  - **Solution Phase (ORNL)**
- **Optimizing gallium concentration (ORNL Neal/Singh, WVU)**
  - **Why add gallium?**
  - **Where is the gallium?**
  - **What is the optimum concentration?**
- **Optimizing hydrogen annealing (ORNL Neal/Singh, WVU)**
  - **Why is a hydrogen anneal necessary?**
  - **What is the optimum process? Does the process change for a ceramic body?**

Combustion Synthesis Time Sequence





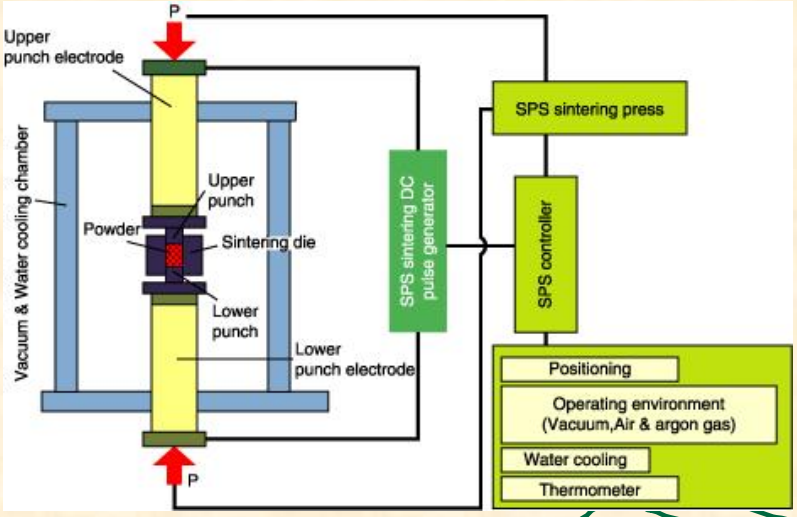


Densification of Precursor Powders

- **Pursuing three methods of densification**
  - **Hot-pressing (ORNL equipment available)**
    - **Uniaxial**
    - **Isostatic**
  - **Vacuum sintering (ORNL equipment available)**
  - **Spark plasma sintering**
    - **UC-Davis has only R&D equipment available**
    - **Only method to successfully densify non-cubic material into transparent body**

ORNL Uniaxial Hot Press

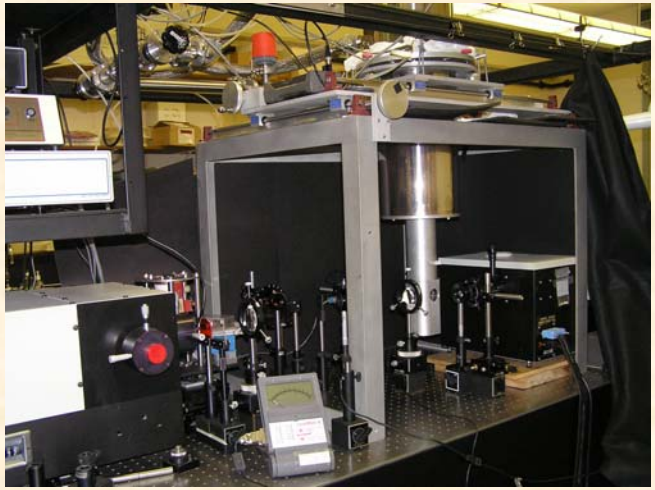
Spark Plasma Sintering Equipment



Characterization of Powders and Densified Bodies

- **Screening techniques**
  - **Photoluminescence (PL) at ORNL**
  - **Alpha and pulsed x-ray excitation at ORNL**
- **Detailed characterization techniques**
  - **Temperature-controlled PL (excitation/emission), EPR, and Raman at WVU**
  - **SEM/TEM/EDS/XRD at ORNL and UC-Davis**
  - **Light output, decay time, timing resolution at ORNL**

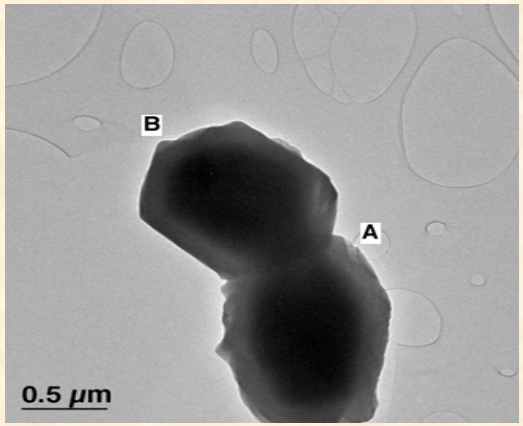
WVU Variable-Temperature (4.2–300 K) PL System

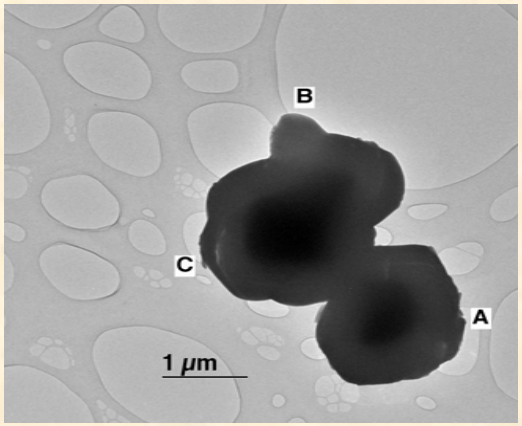


WVU Bruker EMX X-band EPR System (5–350 K)

TEM/EDS and X-ray Diffraction Case Study of Four “Standard”,Urea Precipitation, and Solution Phase ZnO:Ga Powders

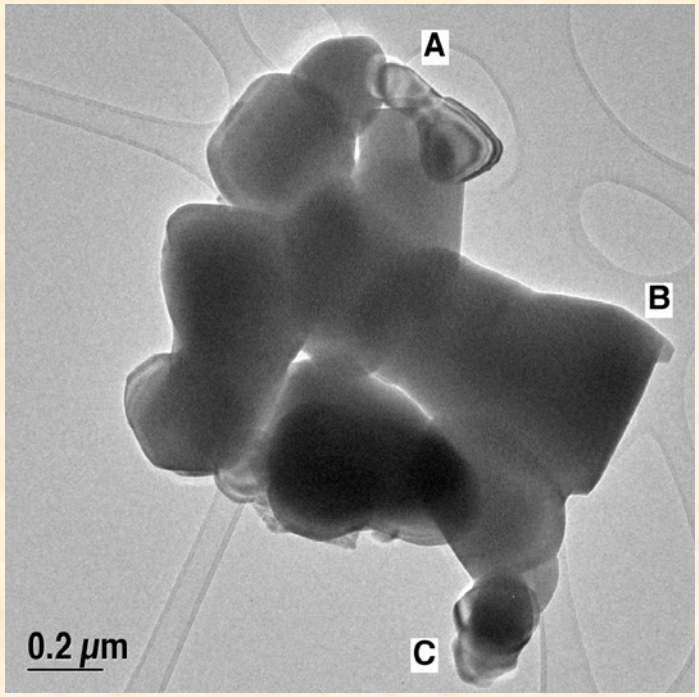
Sample #1: ZnO:Ga WL1201

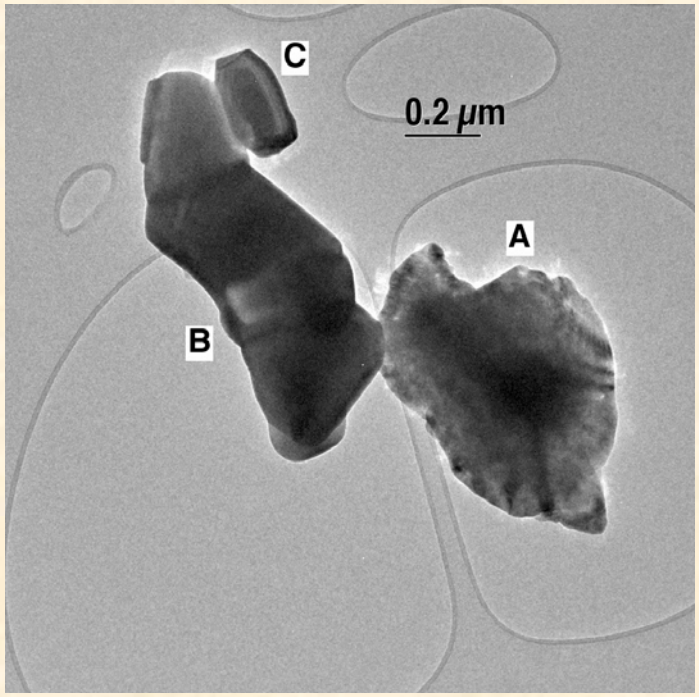




- **The particles size ranges from 0.5-12 microns.**
- **Most of the particles are polycrystalline.**
- **The concentration of gallium varies.**
- **The range of the Ga:Zn ratio: 0.02-0.04.**

Sample #2: NE Tech



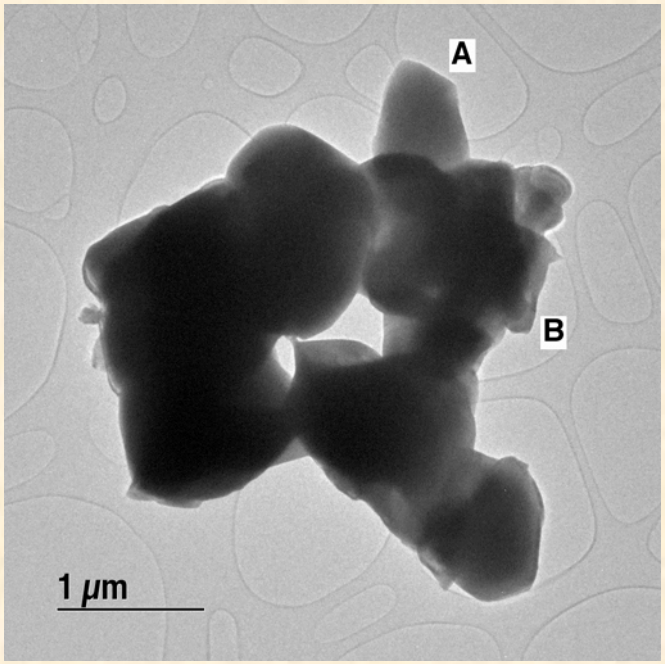


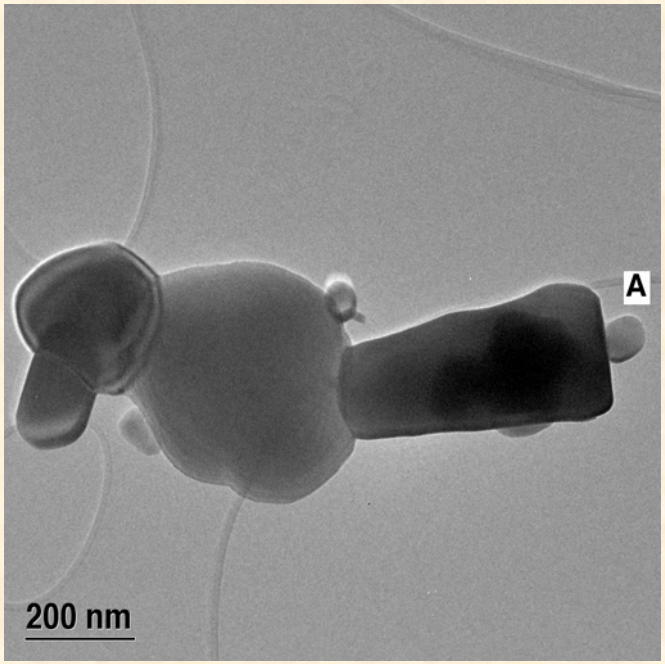
- A, No detectable Ga
- B, No detectable Ga
- C, Ga/Zn= 0.04, Si/Zn = 0.04

- A, No detectable Ga
- B, No detectable Ga
- C, Ga/Zn < 0.01

- Most of the particles are polycrystalline.
- Particle size is in the micron range.
- The Ga concentration varies from non-detectable to 0.02-0.04.
- Silica is present along with the Ga.

Sample #3: ZnO(Ga) LBL 3518



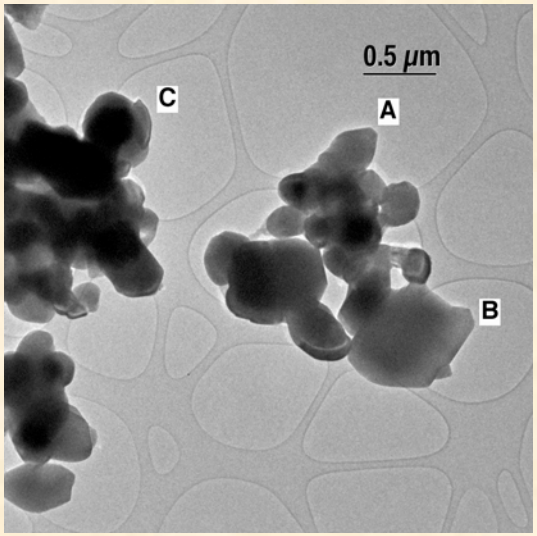


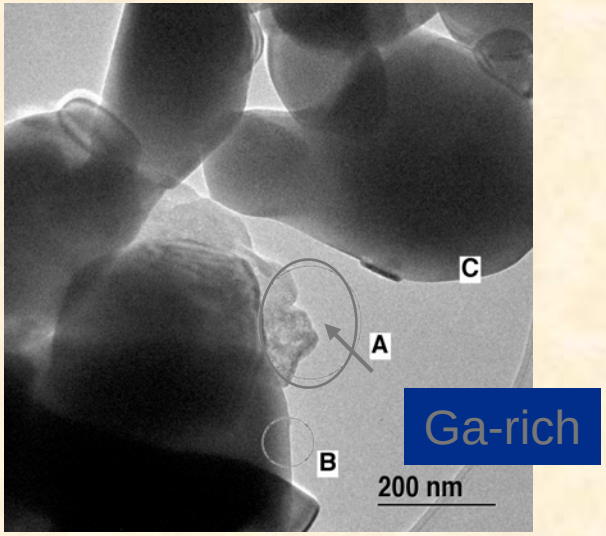
- A, Ga/Zn < 0.01
- B, Ga/Zn= 0.01, Si/Zn = 0.02

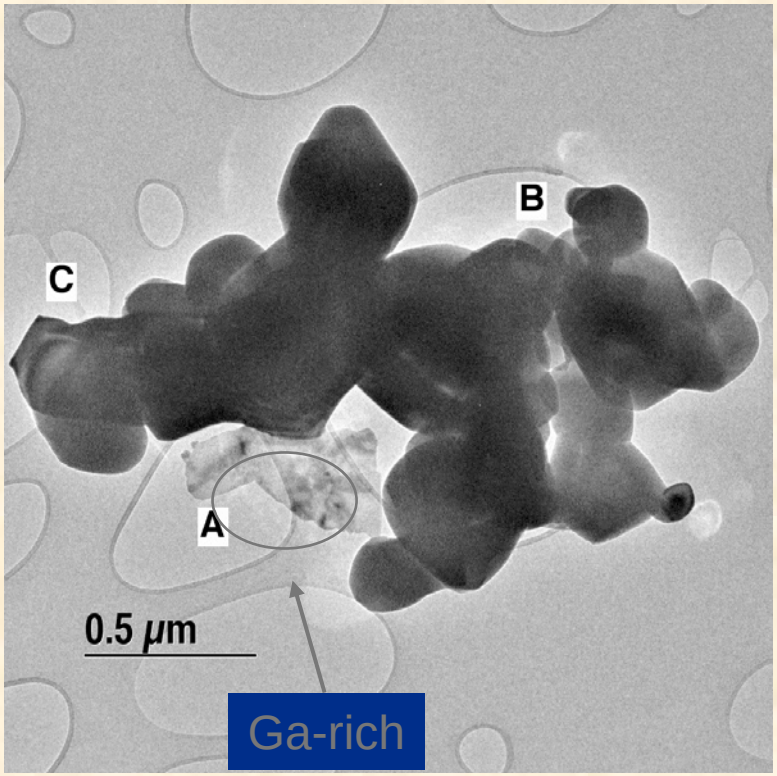
- A, No detectable Ga
- B, Ga/Zn= 0.01, Si/Zn = 0.02, Sn
- C, No detectable Ga, Si/Zn = 0.03

- Most of the particles are polycrystalline.
- The particle size is in the micron range.
- Gallium concentration varies from non-detectable to 0.01.
- Silicon is a major impurity.
- Tin is present in some of the particles.

Sample #4, GK31: Two phases

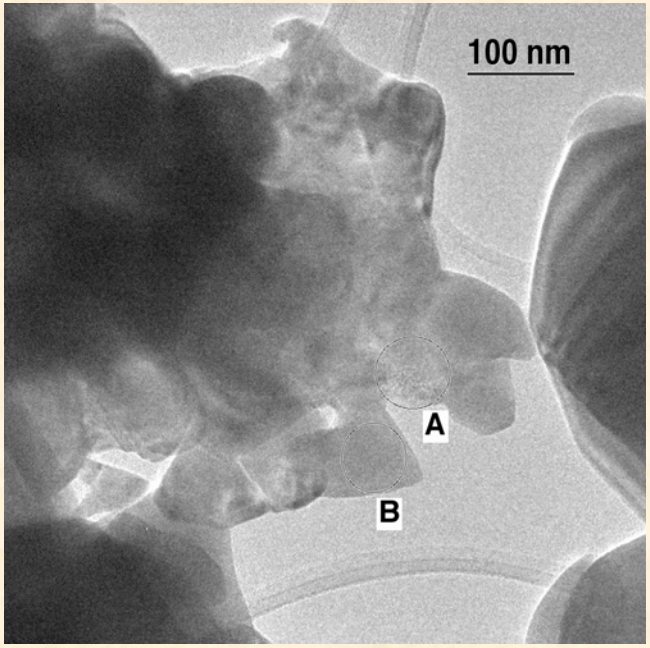


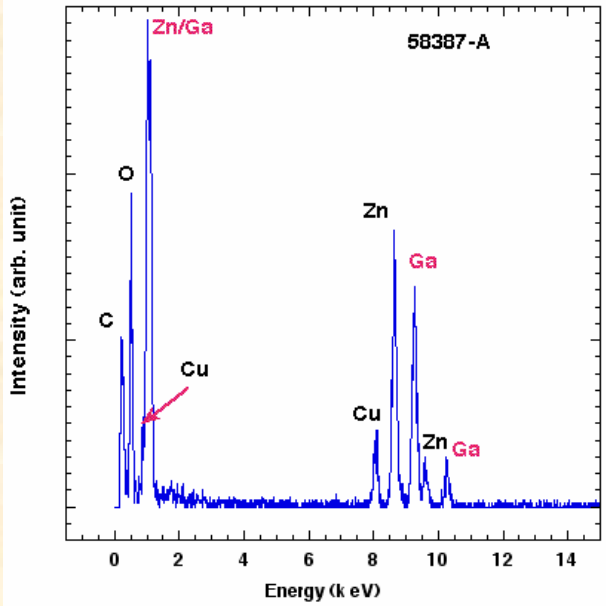


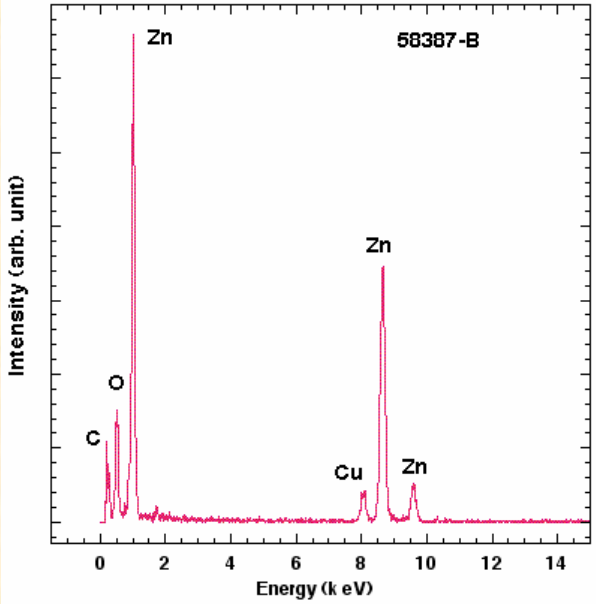


- **This sample has two distinctive phases.**
- **Large polycrystalline particles are zinc oxide with no detectable Ga.**
- **Thin flakes are rich in Ga (circled out).**
- **Most of the particles are polycrystalline.**

Sample #4: High Ga second phase confirmed

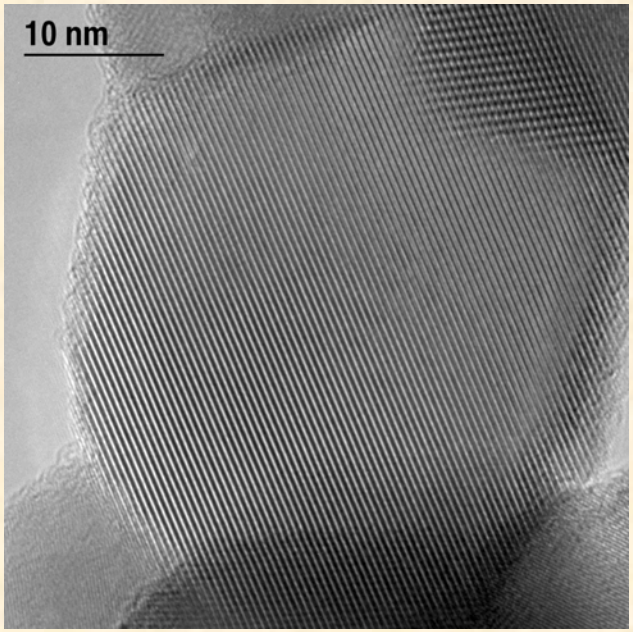


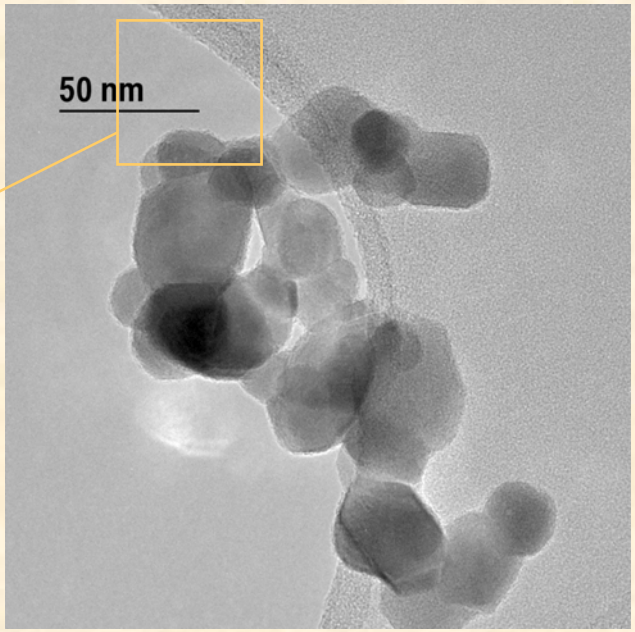




- **EDS analysis indicates that Particle A has about 1:1 Zn:Ga atomic ratio.**
- **No Ga is detect from Particle B.**

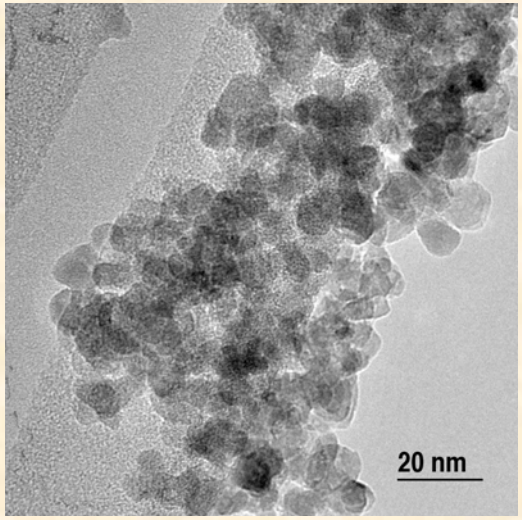
Urea Precipitation: ZnO with 0.04 % Ga #1

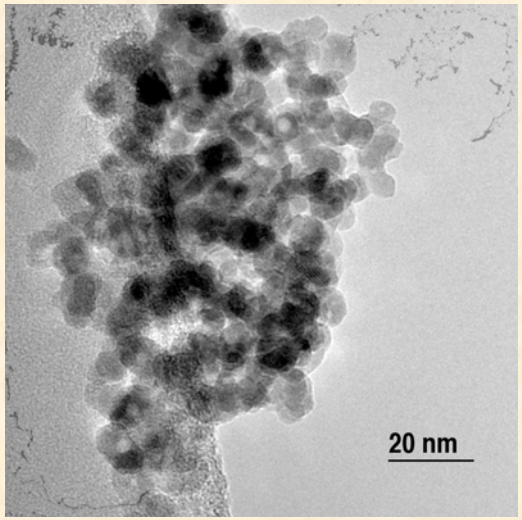


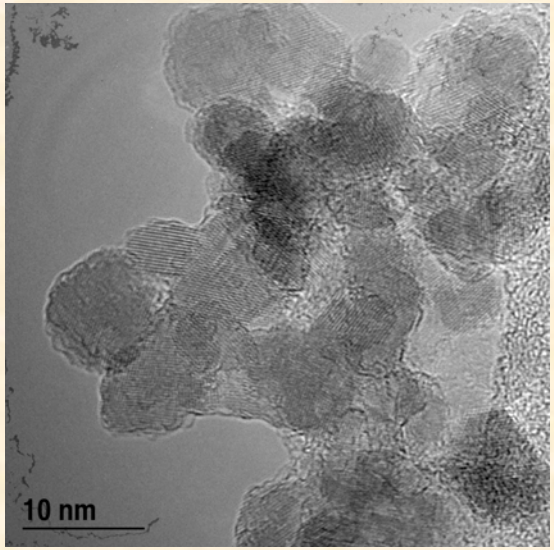


- **A closer view of the small particles with a composition of $ZnGa_{1.9}O_{3.4}$**
- **The small particles are mostly polycrystalline. The majority are attached to the surface of the larger ZnO particles.**
- **Chemical composition was from standard-less quantitative EDS analysis (error +/-10%)**

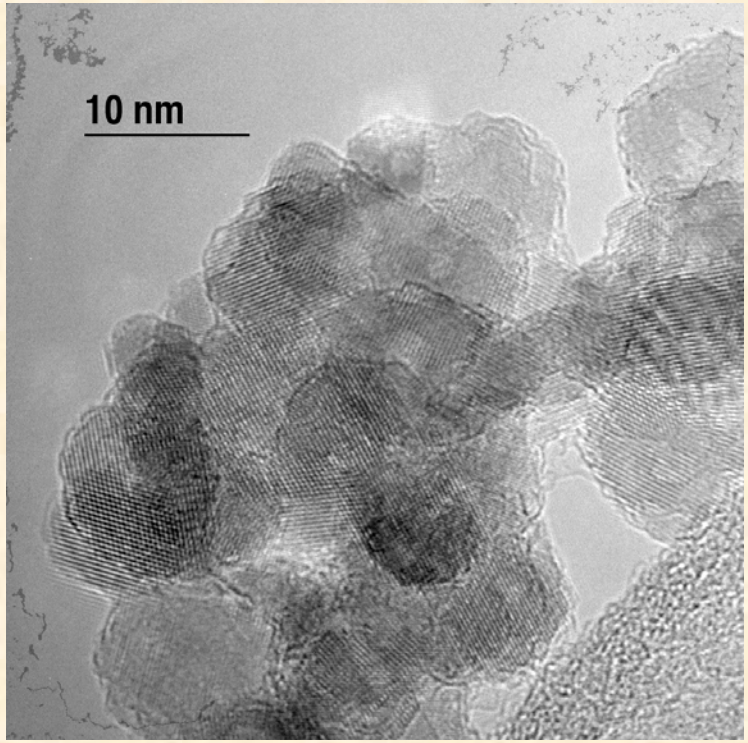
Solution Phase ZnO(Ga)



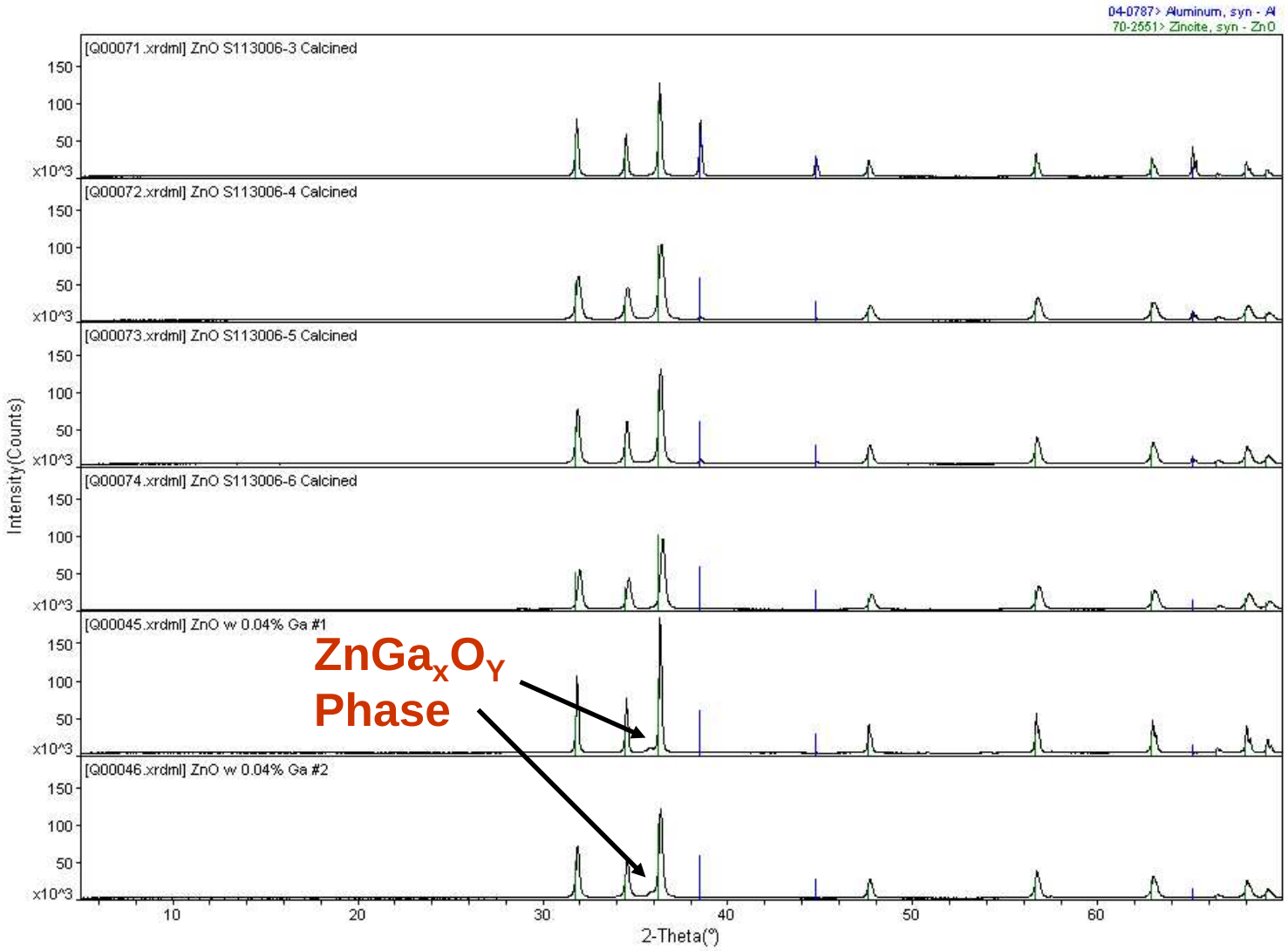




- **Particle size ranges from 5 to 12 nm.**
- **All particles are crystalline.**
- **If there is any Ga, it is at the detection limit level.**



X-ray Diffraction Results for Combustion and Urea Precipitation Samples

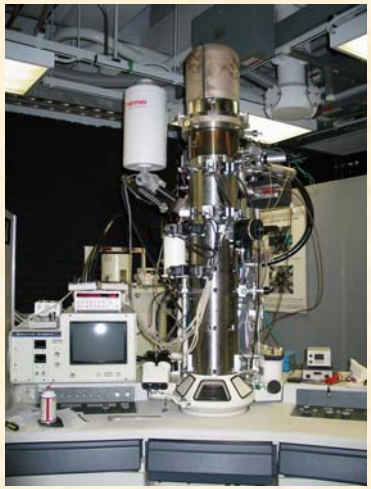


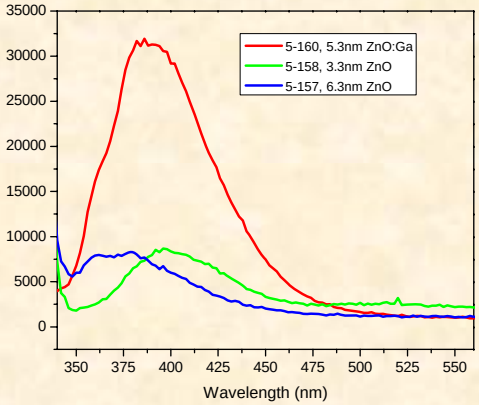
Conclusions from Case Study

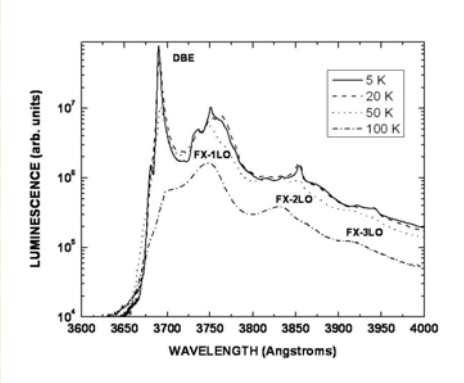
- **Ga is distributed in the samples in different ways**
  - **$ZnGa_XO_Y$ phase in ORNL urea precipitation sample found in XRD and TEM results**
  - **Zn/Ga ratio varies from sample to sample**
  - **Si impurities in some samples**
- **Looking at solution phase nanoparticles with TEM/EDS**
- **No clear trends as to what leads to the best scintillator…still more work to do**

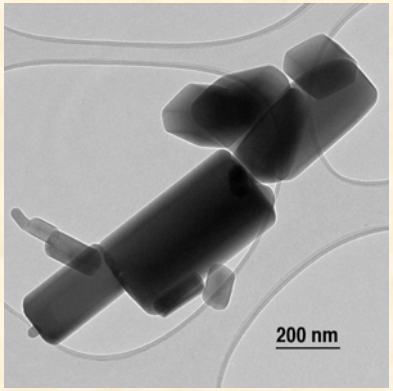
What next?

- **Synthesizing powders using urea precipitation, combustion, and solution phase synthesis techniques. Looking for control of size, shape, doping, and chemical and phase purity**
- **Characterizing powders with XRD, TEM/SEM/EDS, PL, and alpha excitation**
- **Beginning gallium and hydrogen annealing optimization studies**
- **Densification using uniaxial hot pressing and spark plasma sintering**







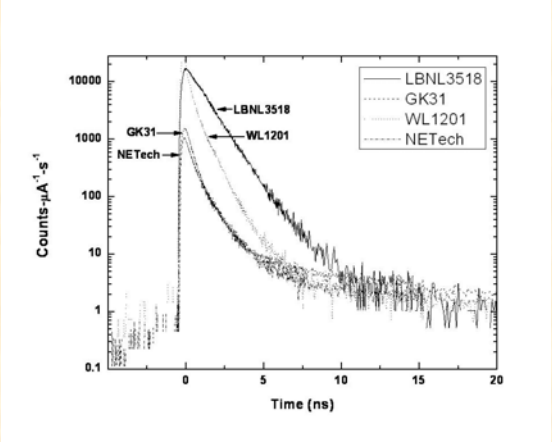


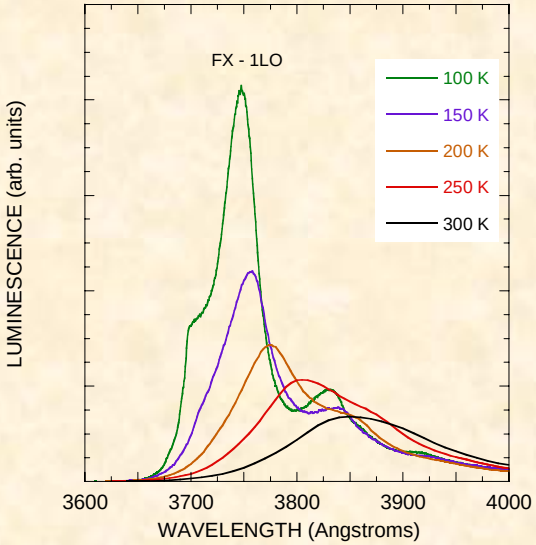
Budget Summary

- **Requested funding**
  - **FY07 $570K**
  - **FY08 $600K**
  - **FY09 $600K**
- **Funding:**
  - **ORNL Physics Postdoc (characterization)**
  - **ORNL Chemistry Postdoc (synthesis)**
  - **Contract with WVU (characterization)**
  - **Contract with UC-Davis (synthesis and densification)**
  - **UT undergraduate-DOE HERE Program (characterization)**
- **Funds expended to date**
  - **$181K**
  - **Does not accurately reflect charges from (2) post-docs, WVU contract, UC-Davis contract**

Summary

- **Supports ORNL neutron imaging system**
  - **Shorter measurement times**
  - **Material ID**
  - **Fission density mapping**
- **Optimizes fastest known inorganic scintillator**
- **Builds knowledge base for transparent polycrystalline ceramic scintillators and extends available materials to include non-cubic scintillators**





Transparent HAp sintered body fabricated by SPS. A hexagonal (NOT CUBIC) crystal

Translucent ZnO:Ga formed by hot pressing at ORNL

Transparent Lu_2O_3:Eu formed by hot pressing at ORNL

Summary of FY07 Technical Deliverables

Task	Due Date	Description	Status
Synthesis of ultra-pure ZnO:Ga powder samples	3/31/07	Internal report on synthesis of ultra-pure ZnO:Ga samples.	In progress
Characterization of ZnO:Ga powder samples	9/30/07	Internal report on characterization of samples.	In progress
Conduct hydrogen annealing studies of powders	9/30/07	Internal report on hydrogen anneal studies.	Commencing
Conduct hot pressing with sacrificial sintering agents	9/30/07	Internal report on hot pressing studies.	Start April 07
Characterization of ZnO:Ga densified bodies	9/30/07	Internal report on characterization of samples.	Start April 07
Identify and establish collaborative effort with a U.S. university to conduct pulsed electric current sintering studies	9/30/07	Internal report on initial pulsed electric current sintering efforts.	Start April 07
All	9/30/07	Document the methodology for synthesis of ultra-pure ZnO:Ga samples and their subsequent densification and characterization in a report to NA-22.	Start August 07