Spark Plasma Sintering: A Novel Procedure for Developing Ceramic Scintillators

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Abstract: Spark Plasma Sintering, or SPS, is a rapid ceramic pressing technique that applies an electric current to quickly heat powdered samples to extreme temperatures, while under a high pressure. Ceramic materials mimic the material properties of their crystalline counterparts. SPS allows the ceramic materials to undergo enhanced densification, which leads to better homogeneity within the samples. This improved densification (>98%) allows the ceramic to have very similar properties as the crystalline counterpart with a higher throughput. With an average growth time of three weeks for bridgeman growth SPS processing is only a fraction of the time. Transparent complex oxide is of particular interest as the low transmission leads to nonradiative centers in optical devicfes. We report the fabrication of ceramic $Lu_2Hf_2O_7$ and $La_2Hf_2O_7$ by spark plasma sintering as a radiation detector.

Introduction: Single crystal scintillators have become one of the most common materials used in technologies that utilize radiation detectors. As the need arise for improved detectors, research into single crystal scintillators has nearly reached its limit. Ceramics provide many benefits over traditional crystal scintillators and have emerged as a promising new scintillator production process. The most ideal process for consolidation powders into a ceramic is spark plasma sintering, shown below. The process is based on the electrical spark discharge phenomenon whereas a high energy, low voltage spark pulse current generates a spark plasma at high temperature resulting in optimum thermal and electrolytic diffusion.

Spark Plasma Sintering Tool

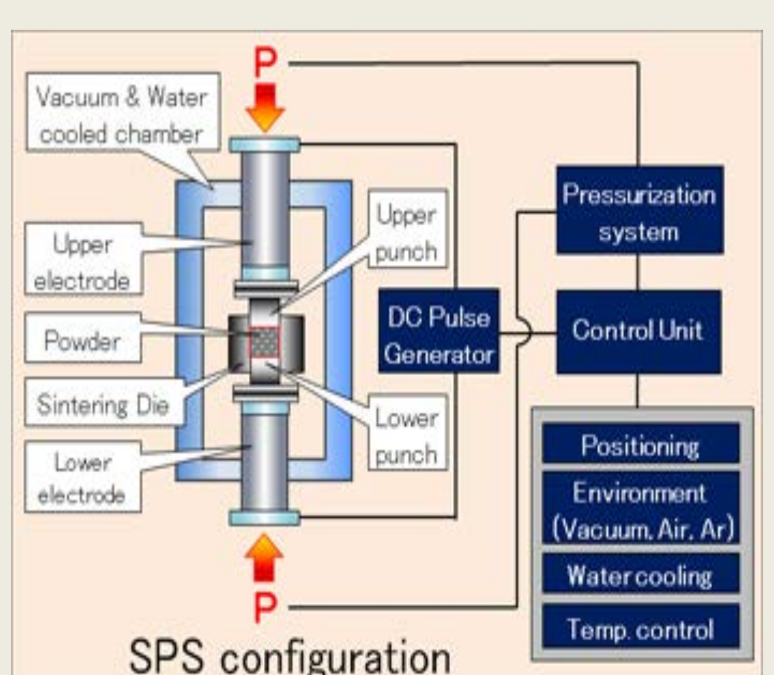
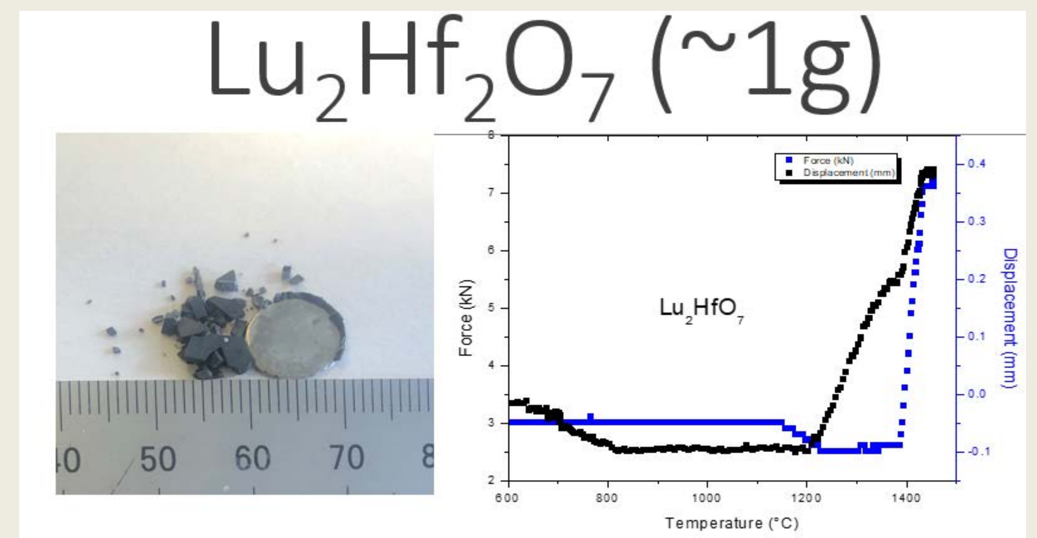
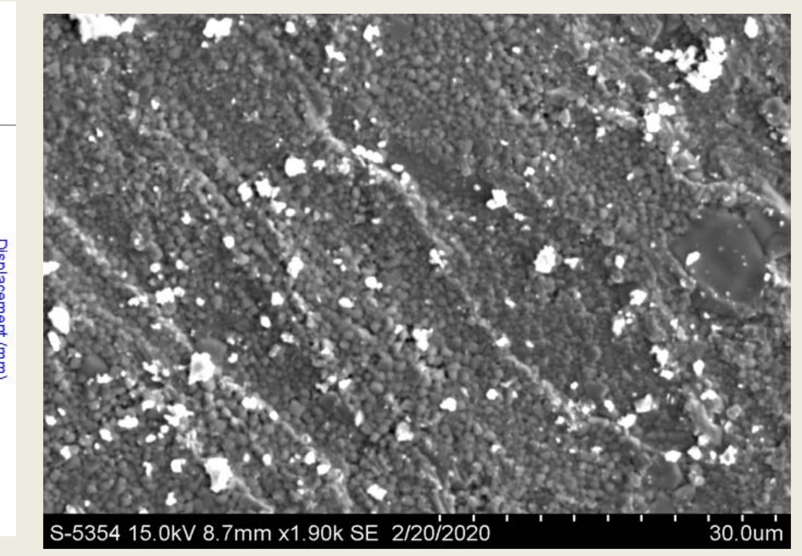


Diagram of the SPS Machine Layout

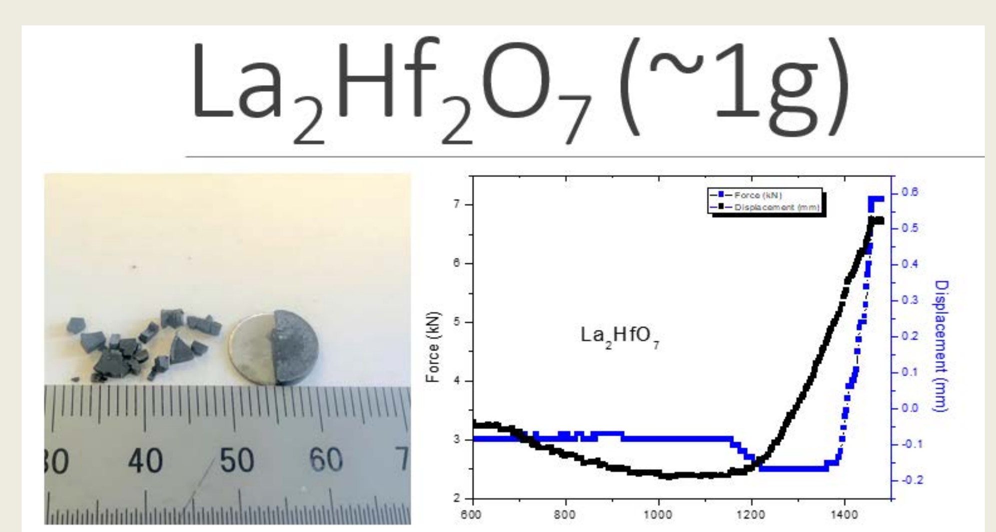


Sample and Sintering Diagram for $Lu_2Hf_2O_7$

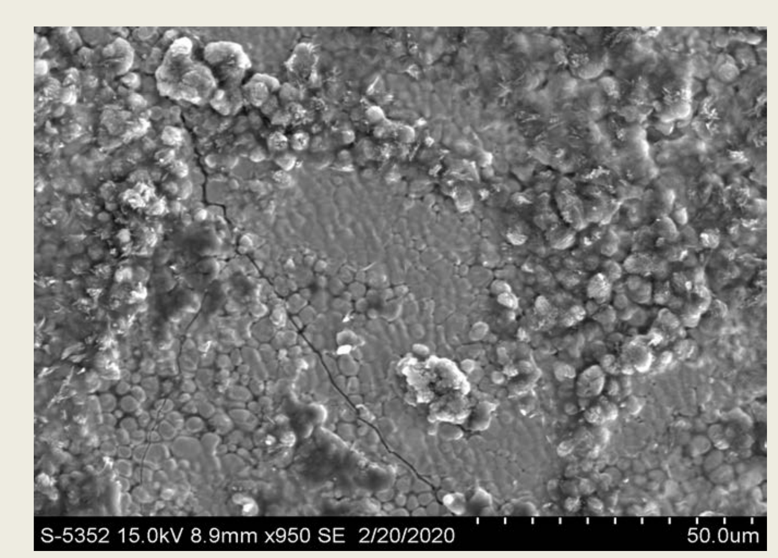


SEM scan of $Lu_2Hf_2O_7$ At 30 μm

Spark Plasma Sintering Conditions:
- Die Size: 10 mm OD
- Pressure: 7.1 kN
- Atmosphere: 90 MPa
- Press time 45 min
- Run Sequence:
  - 700 ℃ with 5 min soak
  - 1450 ℃ with 45 min soak



Sample and Sintering Diagram for $La_2Hf_2O_7$



SEM scan of $La_2Hf_2O_7$ At 50 μm

Spark Plasma Sintering Conditions:
- Die Size: 10 mm OD
- Pressure: 7.1 kN
- Atmosphere: 90 MPa
- Press time 55 min
- Run Sequence:
  - 700 ℃ with 5 min soak
  - 1475 ℃ with 10 min soak
  - 1500 ℃ with 45 min soak

Feedstock Powder Before Sintering

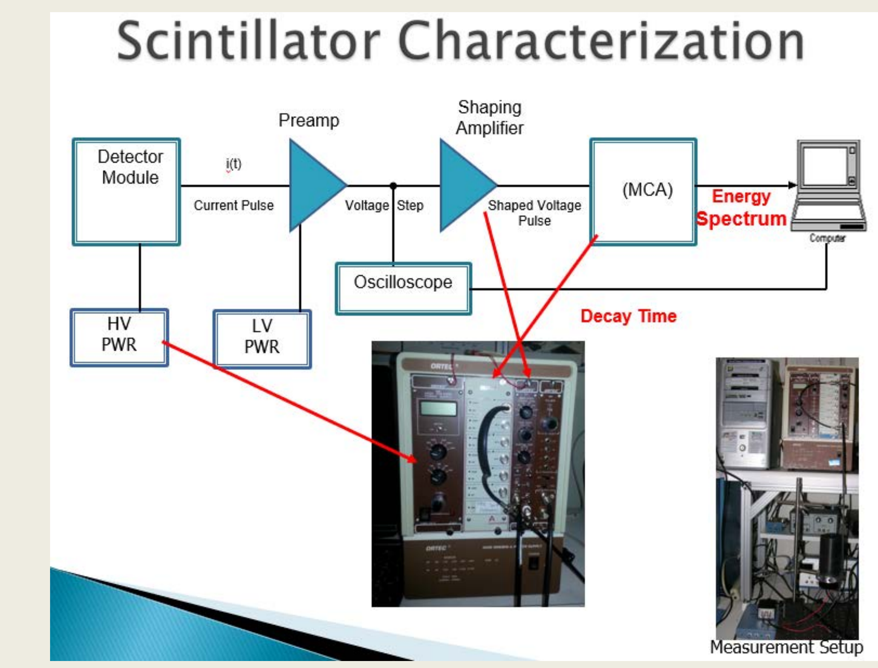


Diagram of light yield detection

Experimental: Powered samples of Lu2Hf2O7 and La2Hf2O7 were proceed at Illinois Institute of Technology. Approximately 1 g of feedstock was used for Spark Plasma Sintering. Scintillation characterization is done using a ^{137}Cs γ source, the setup for γ-ray testing consisted of a Hamamatsu R6231-100 PMT biased at 900V, connected to an Ortec 113 preamplifier, an Ortec 671 spectroscopic amplifier, and a Canberra MP2 multichannel amplifier. Polished samples will be optically coupled to the PMT window using Saint Gobain optical grease. A Spectralon plastic dome will be used as a reflector to increase light collection of diffuse reflections.

Summary: From the content of the received feedstock 10 mm OD sample where produced with a 1mm thickness. Molybdenum foil was used on the punch faces to reduce potential carbon contamination from the graphite die. Both samples came out dark and were cracked during sintering with the lanthanum sample being less cracked. It will take a few sample run with additional material to determine the optimal sintering condition. DSC/TGA measurements were conducted in an argon environment within the range of the system at Fisk University. No change in enthalpy was observed which implies both samples are very stable within the TGA range. After the DSC/TGA run the samples were observed to be light grey in color which gives credence to the need of post annealing to improve the clarity. Both the as-pressed and DSC/TGA "treated" sample did not fluoresce with UV flood exposure which typically indicates poor scintillation.

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