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Investigation of Microcalorimeter Absorber Performance

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Abstract

This work seeks to expand the performance of microcalorimeter gamma-ray detectors such that they are applicable to the wide array of radiation detection needs for nuclear material characterization. We investigated the microstructure of existing bulk tin used to create photon absorbers for microcalorimeter detectors designed by the National Institute of Standards and Technology (NIST) and Los Alamos National Laboratory (LANL). Using this analysis, which included X-ray diffraction (XRD) and scanning electron microscopy (SEM), samples of tin absorbers that provided superior performance were compared to those that provided inferior performance. From the analysis we hypothesize that the crystal lattice orientation and grain size may contribute to the change in performance. We are seeking a fabrication process that will produce tin absorbers with what we hypothesize are good microstructure properties.

Introduction

Microcalorimeter detectors have shown the capability to provide ultra-high energy resolution for measurements of X-rays and gamma rays [1-4]. These detectors provide an improved capability, compared to detectors with worse resolution, to characterize radioactive sources that have a complex photon spectrum because more photopeaks will be resolved. In many fields that deal with nuclear material, a fieldable microcalorimeter could provide new capabilities and methods for material characterization. The work presented in this paper is aimed at improving the performance and scale of spectrometers based on microcalorimeter arrays.

Some current limitations are related to the microcalorimeter fabrication process, which involves hand-assembly of an array of gamma-ray absorbers to an array of transition edge sensors (TES). The TESs are built on a fragile membrane to maximize the thermal transfer from the absorber to the TES and minimize thermal transfer to the perimeter of the array. Additionally, the absorbers are attached to the TESs using SU-8 posts and stycast epoxy. These metal/nonmetal interfaces have a low thermal conductance when compared with a metal/metal interface, where direct coupling of electrons can take place rather than an electron phonon coupling [5]. This hand assembly process makes reliability and scalability to larger detector arrays challenging. Because the volumes of individual absorbers are small (~ 1 mm^3) compared to the active volumes of other radiation detectors, such as high purity germanium (HPGe) or NaI(Tl), arrays with hundreds of elements or more are required to achieve a reasonable detection efficiency. The hand-assembly can also cause some pixels to produce sub-par resolution values and poor pulse timing

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properties compared to other pixels. The microstructure of the absorber may also play a role in performance but knowledge of how performance is affected by varying microstructures is lacking.

To address the limitations caused by hand assembly, we are studying a microfabrication processes that will allow for fabrication of large pixel arrays without hand assembly. First however, we sought to understand the microstructure properties of different tin samples that produced varying performance when used as absorbers. This paper discusses the techniques used for analysis of bulk cold rolled tin absorber materials and attempts to better understand absorber microstructure as it relates to the radiation detection performance.

Absorber Analysis

We analyzed the microstructure of bulk tin samples that were used to fabricate absorbers for NIST/LANL microcalorimeter gamma-ray detectors. The samples were provided by three suppliers and the absorbers fabricated from each exhibited performance differences in energy resolution and pulse timing. Tin from Supplier 1 produced much better energy resolution than the tin from Suppliers 2 and 3. The tin from Supplier 2 also provided better energy resolution than the tin from Supplier 3. Properties of the tin samples are given in Table 1. The supplied thickness of the tin is listed as it was received by NIST and may have been rolled by the supplier to achieve the supplied thickness. The given rolled thickness is the thickness of the tin after an additional rolling process performed at NIST to achieve 0.4 mm thickness. The techniques used for microstructure analysis were XRD and SEM.

Table 1. Properties of tin absorber samples used in analysis

Supplier	Purity (%)	Supplied Thickness (mm)	Rolled Thickness (mm)
1	99.995	0.5	0.4
2	99.995	0.38	N/A
3	99.9	0.8	0.4

Methods

A Bruker D2 Phaser XRD was used to analyze the microstructure of the Sn samples. A Cu K-α X-ray source was used and a wavelength of 1.5406 □ was used to calculate the crystallite size using the Scherrer equation. A Hitachi S-4800 scanning electron microscope was used for high resolution imaging at 1 keV operating voltage.

Results

The XRD analysis showed that the tin from different suppliers had different texture or preferred orientation in their microstructure. XRD spectra are shown for the three samples in Figures 1-3. We found that the Supplier 1 tin has a strong preferred orientation of (211), with a much smaller composition of other orientations such as (111), (101), and (321). Crystallite sizes for sample orientation are shown in Table 2. The (211) crystallite size was larger than 100 nm whereas the other orientations were between 15-20 nm in size. Both the Supplier 2 and Supplier 3 tin have a significant portion of the lattice oriented as (111). In both tin samples the (111) crystallite size is approximately 17 nm and the samples have a higher percentage of nanocrystallinity. The Supplier 2 tin has a larger composition of other orientations than the Supplier 1 tin, but to a much lesser extent than the Supplier 3 tin.

Table 2. Orientations and crystallite sizes for tin samples

Supplier	Orientation	Crystallite Size (nm)	Grains >100 nm in Sample (%)	Grains <100 nm in Sample (%)
1	(211)	> 100	95	5
	(111)	18		
2	(111)	17	14	86
3	(111)	16-17	64	36
	(211)	> 100		
	(101)	> 100		

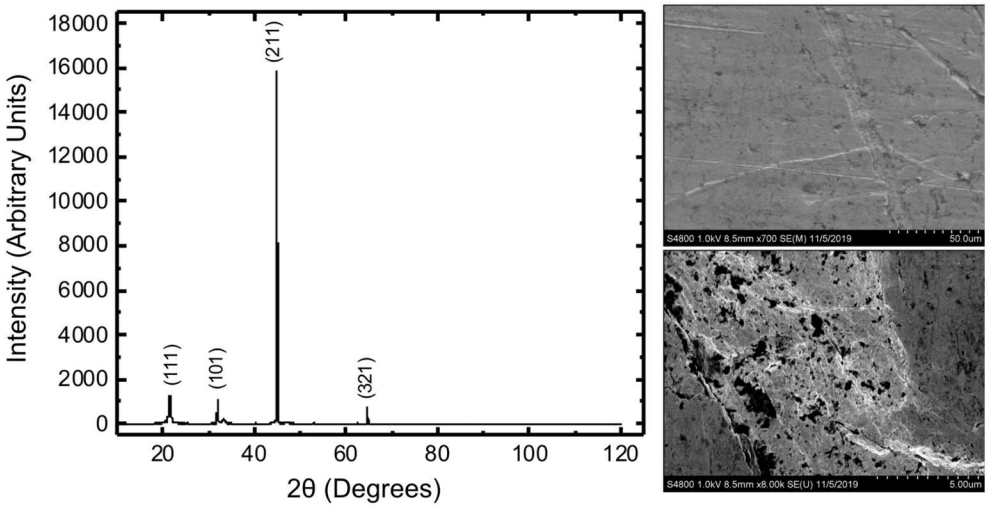

Figure 1. XRD plot (left) and SEM images of Supplier 1 tin using 700X (upper right) and 8000X magnification (lower right).

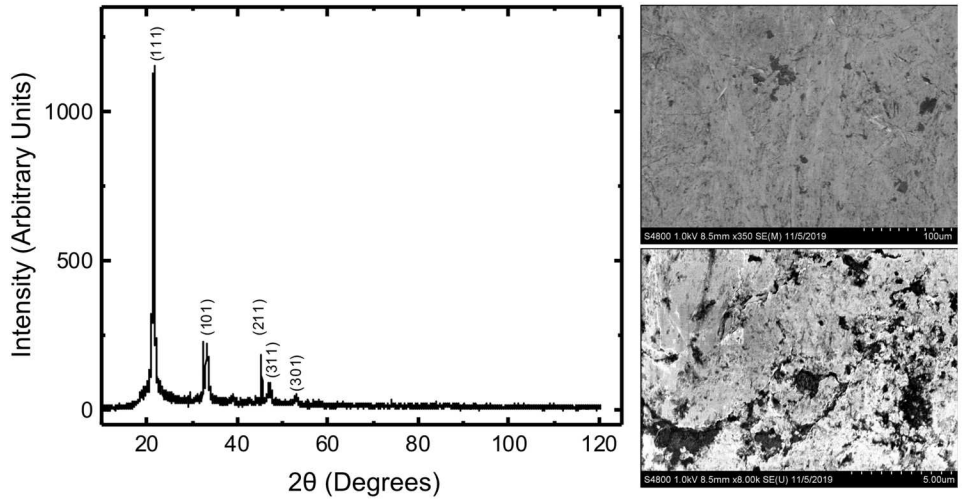

Figure 2. XRD plot (left) and SEM images of Supplier 2 tin using 350X (upper right) and 8000X magnification (lower right).

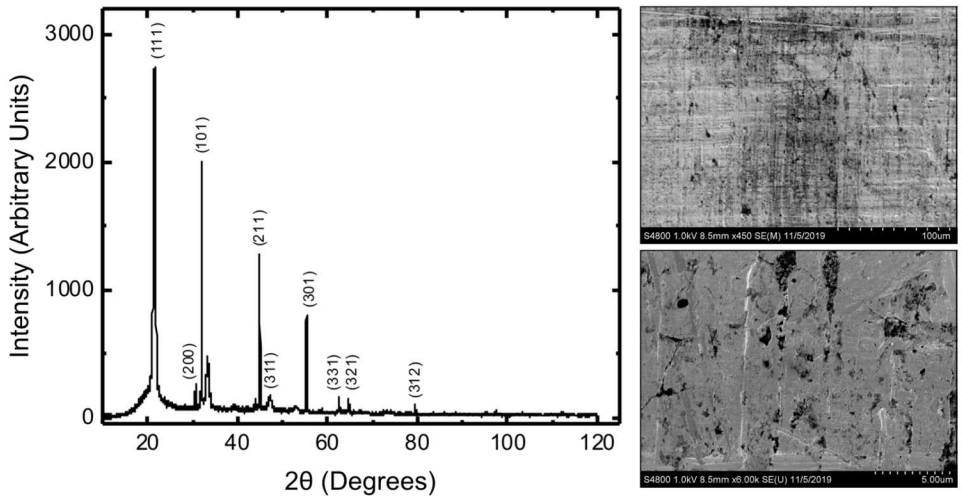

Figure 3. XRD plot (left) and SEM images of Supplier 3 tin using 450X (upper right) and 8000X magnification (lower right).

Discussion

Tin has a body-centered tetragonal lattice structure. In this structure, larger spacing exists between atoms in one dimension of a unit cell compared to the perpendicular direction. Because of this difference, the orientation of the lattice structure will cause different spacing between atoms in the direction that the heat generated by a gamma ray in the absorber must travel to reach the TES. Additionally, the crystal orientation can vary the interface thermal conductance as shown by Hopkins et al. [6]. The tin absorbers are attached to SU-8 posts using an epoxy, which attach to the TES structures and Varghese et al. showed that the thermal conductivity across a metal/nonmetal interface varies based on the percentages of the different orientations of the metal [7]. While the work of Varghese et al. focused on metal/ceramic material, the difference in energy coupling across a metal/nonmetal interface varying with the crystal orientation is consistent. The thermal energy must be transferred by either direct coupling between electrons in the tin and phonons in the epoxy and SU-8 or by electron-phonon coupling in the tin followed by phonon-phonon coupling at the tin/epoxy/SU-8 interface. The crystal plane in contact with the nonmetal will vary the coupling and thus the interface thermal conductivity is varied. More experimentation is needed to better understand this interaction for a tin/epoxy/SU-8 system but absorbers with a (211) texture, that were not nanocrystalline, produced the best energy resolution in radiation measurements.

The microstructure also plays a role in how phonons are generated from the energy deposited in the absorber by an X-ray or gamma ray. Excitation breaks the Cooper pairs creating quasiparticles, which create phonons when they recombine. Microstructure properties such as grain size and varying orientations may affect this process and the production of phonons. A fast recombination is desired, such that the pulse created by an X-ray or gamma ray decays faster, allowing the detector to handle a larger count rate.

Summary and Future Work

Our analysis of the bulk tin absorber samples has shown the microstructure properties of tin that produce the best energy resolution when used as a photon absorber in a microcalorimeter. We will look for fabrication processes that produce tin absorbers with the microstructure properties that were found in the best performing bulk tin. The performance of these absorbers will be tested using an existing NIST/LANL

microcalorimeter spectrometer. We are also investigating methods to directly fabricate the absorbers to the TESs. Such a process will eliminate the hand-assembly methods that currently produce performance variation within an array. Such a fabrication method would enable absorbers with desirable microstructure properties to be fabricated in large and uniform arrays, which will help achieve improved detection efficiency and count rate.

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