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MeV Radiography with GLO Transparent Ceramic Scintillator

Prepared for:

2016 R&D 100 Award Entry

LLNL-PROP-692281

Prepared by LLNL under Contract DE-AC52-07NA27344.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory in part under Contract W-7405-Eng-48 and in part under Contract DE-AC52-07NA27344.

MeV Radiography with GLO Transparent Ceramic Scintillator

1. PRODUCT/SERVICES CATEGORIES

A. Title of Entry

MeV Radiography with GLO Transparent Ceramic Scintillator

B. Product Category (Choose one from online list)

Analytical/Test; Imaging Systems/Devices

2. R&D 100 PRODUCT/SERVICE DETAILS

A. Name of primary submitting organization

Lawrence Livermore National Laboratory (LLNL), Livermore, CA

B. Name(s) of co-developing organization(s) if applicable

Nanocerox, Inc., Ann Arbor, MI

Technology Assessment and Transfer, Inc., Millersville, MD

American Isostatic Presses, Inc., Columbus, OH

C. Product/service brand name

GLO Transparent Ceramic Scintillator

D. Price of product/service in U.S. Dollars

$50,000 per 12” x 12” scintillator, same as scintillator glass in this size

E. Short description of the product (MAXIMUM 25 WORDS)

First-of-its-kind GLO transparent ceramic scintillator transforms MeV radiography by providing 7x faster imaging compared to glass scintillators, greatly decreasing x-ray dose and time-to-image.

3. PRODUCT/SERVICE DESCRIPTION

A. What does the product or technology do?

High Energy Radiography employing mega-electron-volt (MeV) x-rays is the most effective method to look inside large, high-density cast metal parts like power plant turbines, complex metal assemblies such as jet engines, as well as to identify defects in welds, for example, during ship-building [1-2]. It is also used to non-destructively look inside of items from antiquity, such as fossilized dinosaur eggs and Roman statues. As imaging technology improves, new applications become feasible, such as non-destructively looking at the three-dimensional (3D) volume of complex objects in real time and enabling 3D reverse engineering without disassembling complex machines. While traditional film and flat panel two-dimensional (2D) radiographs are helpful, 3D reconstructions of the interior of dense objects via x-ray computed tomography (x-ray CT) provides a complete map of internal structures that cannot be obtained in any other way.

X-ray photographic film can provide excellent spatial resolution of ~20 microns (limited by the emulsion grain size); however, film cannot provide CT imagery. Flat panel radiography arrays can acquire many views of a rotating object for CT reconstructions, which can take several hours or even a full day for complex objects. Array spatial resolution is pixel-size limited to ~150 microns, but in practice ~0.5 mm is typical for MeV imaging [1,3]. To achieve high-resolution x-ray CT imaging for large, complex, or dense objects, LLNL previously developed an alternative approach,

a lens-coupled radiography system named CoLOSSIS (Confined Large Optical Scintillator Screen and Imaging System), which was fielded at Pantex (Amarillo, Texas) in 2009 [4, 5]. This system, depicted schematically in Figure 1, consists of a 9 MeV Varian Linatron Bremsstrahlung x-ray source, a rotating platform for the object, a large glass plate scintillator, an imaging lens system, and a visible light CCD with 15-micron pixels. The x-ray source and acquisition geometry limits the spatial resolution of the system to >25 microns

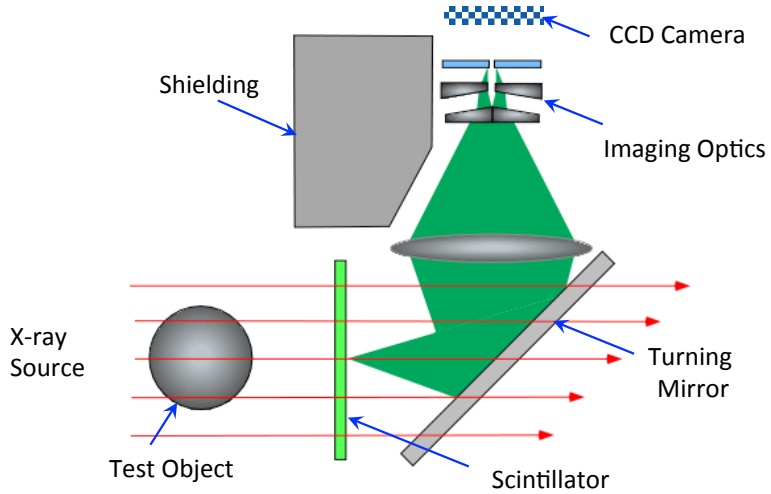


Figure 1. Experimental arrangement for lens-coupled radiography. Collimated x-rays interact with the object, and the image is formed in a free-standing transparent scintillator sheet. The image is reflected by a turning mirror, into imaging optics and then recorded by a high-performance, low-noise CCD camera.

First-of-its-kind $(Gd,Lu,Eu)_2O_3$, or "GLO," transparent ceramic outperforms existing standard scintillator glasses and single crystals in mechanical toughness, uniform material response, and manufacturability into the large, clear sheets needed for radiographic imaging. Transparent ceramics are an emerging class of optical materials, with applications including transparent armor, "unbreakable" windows, missile domes, lenses, laser gain media, and scintillators [6,7]. Transparent ceramics are polycrystalline, monolithic, fully dense optics that offer advantages in low-processing temperatures, ease of fabrication of complex shapes and high-aspect-ratio optics (such as plates), and high, uniform doping, needed for high-performance scintillators. Many efforts to fabricate optically transparent Lu_2O_3 with Eu doping have been made over the years, but ultimately were never able to achieve sufficient transparency for lens-coupled radiography

applications [8,9]. This is because Eu has limited solubility in Lu_2O_3, due to the mismatch of the ionic radii of Eu (95 pm) with Lu (86 pm), so above the 1% Eu doping level, Eu-rich secondary phases form at the grain boundaries of Lu_2O_3. We discovered that this can be mitigated by the addition of 10% or more Gd (ionic radius of 94 pm), substituting for Lu, whereupon high-transparency ceramics, with high Eu doping of 5%, and without secondary phases, may be fabricated [10,11].

The GLO transparent ceramic scintillator dramatically increases high-energy radiography throughput by providing 7x faster imaging with 9 MeV x-rays than glass, and decreasing the x-ray dose required to obtain detailed imagery. GLO outperforms the standard IQI glass scintillator, due to its 2.8x higher light yield and 2.6x higher x-ray stopping (GLO's density is 9.1 g/cm^3 and its effective atomic number, Z_{eff}, is 68). Furthermore, large field-of-view (12" x 12") GLO transparent ceramics, available for the first time in 2016, provide spatial resolution of 59 microns (8 lp/mm) with 9 MeV x-rays, which approaches the geometry-limited resolution for the system.

B. How does the product operate?

Current commercial production of transparent ceramics in plates is not widely available in large plates, although there are two materials being manufactured for transparent armor windows, AlON and $MgAl_2O_4$, neither of which functions effectively as scintillator. The current 12" x 12" GLO transparent scintillator plates fabricated by LLNL, and just now available in 2016, approach the largest contiguous transparent ceramic optics available, of any material. The transparent ceramics fabrication steps used by LLNL entail a specialized process that combines modern nanotechnology with domestic infrastructure for large-scale ceramics fabrication and required

GLO Transparent Ceramic Fabrication Steps

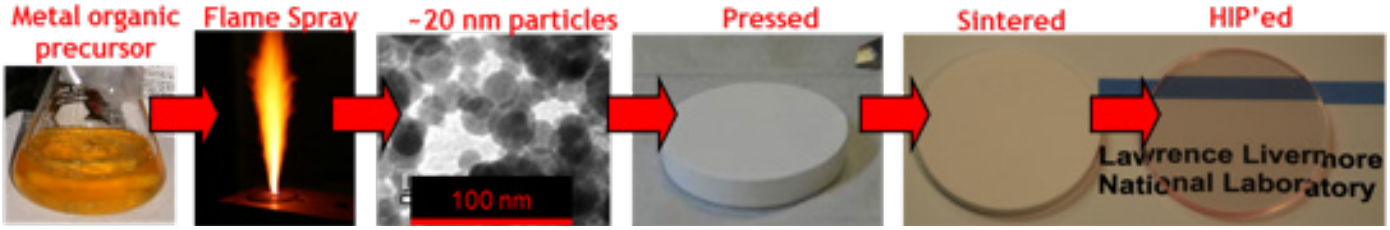
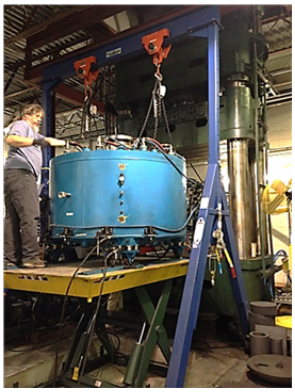


Figure 2. Transparent ceramics are fabricated at LLNL via a unique route employing nanoparticles synthesized in a flame that are then hot pressed, sintered, and then hot-isostatic-pressed to transparency.

six years to develop the precise processing conditions (detailed in Figure 2). Starting with stoichiometric mixed metal oxide nanonparticles synthesized via flame spray pyrolysis, a ceramic "green body" is formed via uniaxial hot pressing, which is then sintered at 1600° C, followed by hot isostatic pressing. For scaleup to 10" round and 12" x 12" square GLO scintillators (Figure 3), LLNL collaborates with three industrial partners: (1) Nanocerox, Ann Arbor, Michigan, for large-scale flame spray pyrolysis powder production; (2) Technology Assessment and Transfer (TA&T), near Baltimore, Maryland, for uniaxial hot-pressing of green bodies; and (3) American Isostatic Presses (AIP), Columbus, Ohio, for hot isostatic pressing. The close multi-year collaboration between LLNL and these three companies was critical to the success of this work. The intellectual property of the GLO scintillator is protected with our US Patent 9,238,773 [10], and the methodology is described in [11]. Multiple large GLO scintillators are shown in Figure 4.

NANOPOWDER SYNTESIS - NANOCEROX, INC.

HOT - PRESSING - TA&T



HIP - AMERICAN ISOTATIC PRESSES, INC.

Figure 3. (left) Large-scale GLO nanopowder is produced at Nanocerox, Inc. (middle) GLO ceramic green bodies are hot pressed at TA&T, Inc. and (right) then hot-isostatic-pressed at AIP.

Figure 4. Several large GLO scintillators, 6-10" across. At bottom, the same scintillator is shown in room lights, and under UV excitation (at right), showing the red Eu^{3+} fluorescence that GLO produces when it scintillates.

4. PRODUCT COMPARISON

A. Supply a matrix or table showing how the key features of your product compare to existing products or technologies. Use numerical figures to represent performance metrics. For price, and capital and operating costs, use actual dollar amounts or a relative scale to show a comparison.

To compare scintillator candidates, we define here a normalized Effective Light Yield, **ELY (unitless)**, under irradiation with 9 MeV Bremsstrahlung (average energy is 3 MeV). The product of the mass attenuation coefficient, **μ (cm^2/g)**, and the material density, ρ **(g/cm^3)**, is the material stopping power at a given x-ray energy, α **(cm^{-1})**. The light yield, **LY (Photons/MeV)**, is the efficiency of the scintillator for converting ionizing radiation (x-rays) into visible photons.

Effective Light Yield = α × LY

Scintillator	ρ (g/cm^3)	α (cm^{-1}) @ 3 MeV	Light yield (Ph/MeV)	ELY (unitless)
GLO, $Gd_{0.3}Lu_{1.6}Eu_{0.1}O_3$	9.1	0.36	55,000	7.1
CsI(Tl) – available in small sizes only	4.5	0.17	65,000	3.9
$CdWO_4$– available in small sizes only	7.9	0.30	28,000	3.1
IQI Glass	3.8	0.14	20,000	1.0

Table 1. Comparison of scintillators for high energy x-rays. The only scintillators that can be fabricated into large sheets are GLO and IQI glass; the other scintillators are single crystals and are only available in sizes much smaller than needed for 12" field-of-view MeV imaging. The Effective Light Yield for GLO is calculated to be 7.1 x better than that of IQI glass.

B. Describe how your product/service improves upon competitive products or technologies.

Attenuation radiographs as well as computed tomography studies were performed, using the only two large area scintillators available, GLO ceramic and IQI glass, both 2 mm thick. Attenuation radiographs were acquired with a ^{238}U penetrameter, and modulation transfer function (MTF) fits were performed on edge features in order to obtain the spatial resolution. Additional radiographs were acquired with a single-crystal nickel-turbine blade (Table 2), and these experimental results show that the Effective Light Yield of GLO is 7.5x higher than the IQI glass, comparable to the 7.1x calculated in Table 1. The spatial resolution is slightly improved by a factor of 1.2. Computed tomography reconstructions of the turbine blade reveal that for a fixed acquisition time, the higher light yield and resolution of the GLO scintillator provides a crisper, higher-contrast image. Operationally, the higher Effective Light Yield of the GLO scintillator will allow it to collect a high quality CT reconstruction in an hour, instead of a full day. The higher light yield also opens up opportunities for new ways of handling the data and image collection, involving binning and frame rate that will speed up the time-to-image even further. Since the properties of the x-ray source and geometry in 9 MeV lens-coupled radiography systems typically limit the spatial resolution

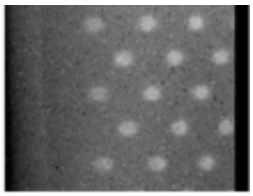
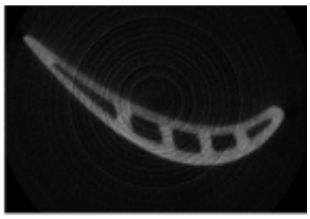
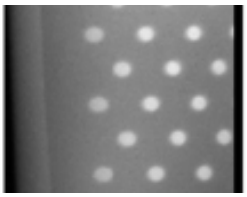
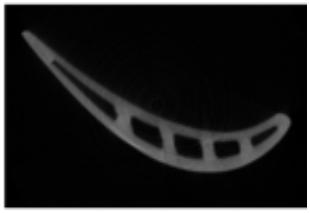
Scintillator	Attenuation Radiograph	CT Reconstruction	Rel. Light Yield	Resolution (microns)
IQI Glass			1	70
GLO			7.5	59

Table 2. ***Comparison of performance of GLO with IQI glass scintillator. (Col. 1)*** *Photos of IQI glass and GLO transparent ceramic scintillators. Images acquired of the turbine blade, with internal structural pillars, each with the same camera exposure time.* ***(Col. 2)*** *Attenuation radiographs showing the internal pillars.* ***(Col. 3)*** *Computed tomography reconstructions indicate improved contrast, resolution, and reduction in ring artifacts for the images acquired with the GLO transparent ceramic scintillator, for the same data acquisition time.*

to >25 microns, the GLO scintillator resolution, at 59 microns, approaches the achievable resolution limit [12]. We have performed additional comparative studies of the GLO and IQI glass scintillators, and while the IQI glass darkens under radiation exposure, the GLO scintillator does not measurably darken even following a 53 MRad dose of 9 MeV x-rays , increasing the usefulness of GLO for high x-ray fluence applications.

C. Describe limitations of your product/service.

GLO is initially vectored to MeV imaging systems that are large-scale and specialized. We have concurrent efforts in keV imaging that have proven that GLO's deployment in smaller systems offers significant advantages [13-14]. GLO is in fact close to being incorporated into some commercial instruments, operating in the keV range. Owing to the proprietary and competitive nature of this business, we are unable to provide the details, but a company is integrating GLO into its keV x-ray imaging devices as a "drop in" replacement for other scintillators because of the spatial resolution enhancement GLO provides in the keV x-ray range.

5. SUMMARY (Maximum 175 words)

The new transparent ceramic GLO scintillator provides dramatically improved throughput for MeV radiography and cone-beam computed tomography. It has been scaled up to 12" x 12" optically transparent sheets that are mechanically tough and radiation hard. GLO offers spatial resolution that is better than the available glass scintillators for lens-coupled radiography, while its combined light yield and stopping power improvement results in >7x greater scanning speed, with higher contrast performance. Currently employed industrial x-ray computed tomography systems for imaging of high-attenuating, high-density components such as rocket motors, engines, turbines, automotive assemblies, and heavy machinery are costly to operate, since they take so long to acquire useful imagery. Implementation of the GLO scintillator will allow an image that used to take a full day to be acquired in one hour, permitting more components to be imaged cost-effectively, leading to rapid manufacturing feedback for quality control and enabling the production of high-reliability components and the development of new capabilities such as real-time CT imaging.

6. CONTACT INFORMATION

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7. AFFIRMATION

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8. LETTER OF ENDORSEMENT

VJ TECHNOLOGIES

May 18th, 2016

To: Dr. Nerine Cherepy <cherepy1@llnl.gov>
LLNL Lead Scientist, SPIE Fellow

Dear Ms. Cherepy,

VJ Technologies (VJT, VJE and VJX) are world leaders in the development of x-ray inspections and NDT solutions with fielded systems worldwide. VJT also provides state-of-the art x-ray inspection services for industrial objects at length scales from 0.005 mm for low-energy microfocus inspections to 0.1-0.2 mm inspection work at 6 or 9 MeV.

VJ Technologies currently operates a multi-source high-energy facility in East Haven Connecticut. The facility includes both 6 and 9 MeV LINAC sources, and each of these x-ray sources can be coupled with Amorphous Silicon panel x-ray detectors or the different linear arrays available for use. The motion capability connected to this facility includes a 4000 lb overall capacity with x-y-z and theta machine controlled operation. Routine DR and CT scanning at this facility includes a wide range of industrial objects from high-value aerospace assemblies, Nuclear Waste Containers and Drums, large industrial objects, and geometry acquisition of automotive assemblies.

VJT is acutely interested in the fielding a system which includes the high-spatial resolution and high-stopping power properties of the GLO scintillator. We currently have customers asking for higher spatial resolution inspections using MeV radiation. Our first application would include detailed scanning of automotive and aerospace objects at t levels of contrast and resolution that have not been demonstrated to-date. Important to our operation, we understand the increased brightness of GLO to enable these inspections to be performed at much increased speeds – adding to our throughput.

We expect addition of a scanner with the properties of the GLO scintillator, configured as a Cone-Beam CT imager will enable new opportunities in x-ray inspection at Medium to Higher energies.

Best Regards,

Stephen Halliwell
General Manager, VJ Inspection Services

Bohemia, NY • East Haven, CT • Littleton, MA • Atlanta, GA • Paris, France • Bangalore, India • Suzhou, China

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