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The characterization of scintillator performance at temperatures up to 400 degrees centigrade

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ABSTRACT

The logging and characterization of geothermal wells requires improved scintillator systems that are capable of operation at temperatures significantly above those commonly encountered in the logging of most conventional oil and gas wells (e.g., temperatures nominally in the range of up to 150 °C). Unfortunately, most of the existing data on the performance of scintillators for radiation detection at elevated temperatures is fragmentary, uncorrelated, and generally limited to relatively low temperatures—in most cases to temperatures well below 200 °C. We have designed a system for characterizing scintillator performance at temperatures extending up to 400 °C under inert atmospheric conditions, and this system is applied here to the determination of scintillator performance at elevated temperatures for a wide range of scintillators including, among others: bismuth germanate, cadmium tungstate, cesium iodide, cesium iodide (Tl), cesium iodide (Na), sodium iodide, sodium iodide (Tl), lutetium oxy-orthosilicate (Ce), zinc tungstate, yttrium aluminum perovskite (Ce), yttrium aluminum garnet (Ce), lutetium aluminum perovskite (Ce), and barium fluoride, strontium iodide (Eu). Most of the scintillator samples exhibited severe degradation in light yield at elevated temperatures. Measurements were terminated at temperatures at which the measured light yield no longer appeared useful. The results of these high-temperature scintillator performance tests are described in detail here. Comparisons of the relative elevated-temperature properties of the various scintillator materials have resulted in the identification of promising scintillator candidates for high-temperature use in geothermal and fossil-fuel well environments.

1. Introduction

The oil and gas industry has a long history of employing nuclear detection (as well as a number of other techniques) to characterize the properties of geologic formations as a function of depth in wells that are drilled for the purpose of fossil fuel extraction [1]. This characterization process, commonly referred to as "well logging", includes the determination of a wide range of formation characteristics—including, among others, the temperature, pressure, permeability, resistivity, a quantity known as the "spontaneous potential", porosity, density, liquid (water) content, natural radioactivity, and chemical composition. In the case of nuclear techniques, gamma ray spectroscopy is used to identify the source of natural radioactivity; neutron techniques are generally used to determine porosity, water content, and density; while combined neutron activation and gamma ray spectroscopy methods are used to identify the elemental composition of non-naturally radioactive elements that are present in the formations.

The nuclear-based well logging "tools" that are presently employed in the oil and gas industry can generally operate in the temperature range of 150 to 175 °C—i.e. in the elevated temperature range characteristic of the down-hole conditions usually present in oil and gas wells. In the case of "logging" or characterizing geothermal wells, however, the down-hole temperatures can reach up to 375 °C for well depths up to 10,000 ft. Accordingly, due to their extreme temperature environments, current nuclear-based oil and gas well logging technology is not directly applicable to the characterization of geothermal wells [2,3].

This work addresses current deficiencies in one-time measurements of wellbore and system parameters at high temperatures [2,3]. Specifically, the present work addresses one facet of the issue of radiation detector performance under geothermal well logging conditions by characterizing the relative performance of

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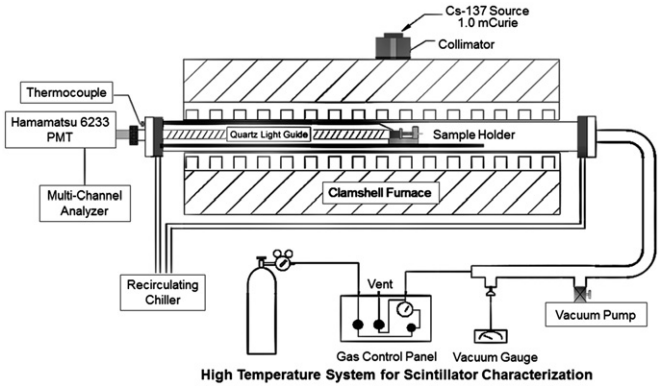

Fig. 1. Schematic diagram of the system used for the characterization of the elevated temperature properties of scintillators. A clamshell Mellen Co. resistance-heated furnace and a Honeywell PS306 temperature controller provide the required variable temperature conditions via a thermocouple located at the sample position. The sample holder assembly is contained in a stainless steel enclosure with water-cooled end flanges that is filled with Argon during the measurements. Light from the scintillator is coupled to a quartz light pipe and transmitted to a photomultiplier tube that is external to the furnace. Excitation of the scintillators is provided by 662 keV gamma rays from a collimated 1.0 mCurie source.

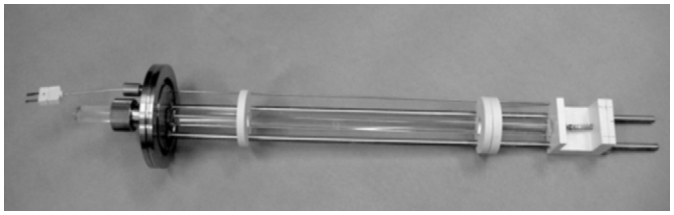

Fig. 2. View of the scintillator sample holder assembly. The white rings are boron nitride (BN) spacers used to center and align the insert in the stainless steel enclosure. The white sample holder assembly located near the end of the two stainless steel support rods consists of a BN body with a moveable spring-loaded platen that holds the scintillator sample flat against the end of the quartz light guide.

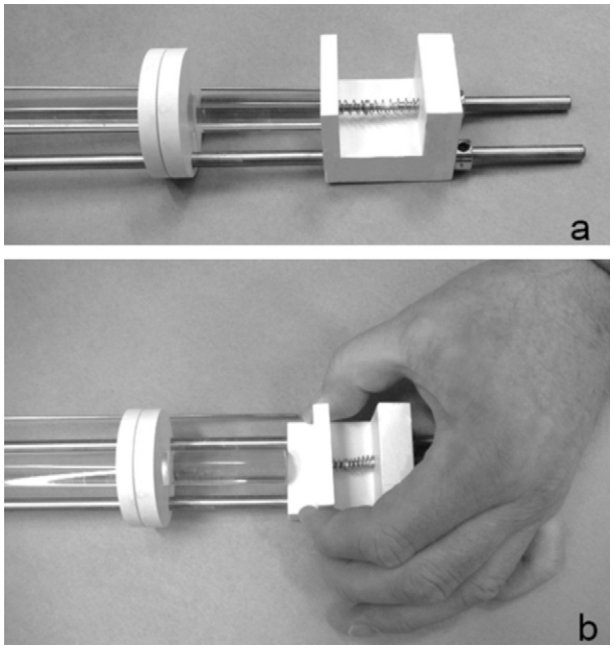

Fig. 3. Close-up view of the boron nitride sample holder assembly: (a) with the spring-loaded BN platen fully forward against the quartz light guide, and (b) with the spring loaded platen withdrawn so that the assembly is ready for the insertion of a scintillator crystal.

a wide range of gamma ray scintillator materials at temperatures up to 400 °C. The scintillators investigated here primarily represent a range of commercially available gamma ray detector materials, and many of these scintillators were, in fact, either purchased directly from commercial scintillator manufacturer/suppliers—or they were donated by such suppliers for use in the subject present elevated-temperature scintillator performance studies. In a few cases, less-common or highly hygroscopic scintillators that were difficult to obtain or ship in an un-encapsulated or "un-canned" state were grown in single crystal form and fabricated on-site in the Center for Radiation Detection Materials and Systems at the Oak Ridge National Laboratory.

Table 1
Summary of tested materials and respective sources and sizes.

Material	Source	Crystal size (mm)
$Bi_4Ge_3O_{12}$ (BGO)	Hilger crystals	$10\times10\times10$
NaI:Tl	Hilger crystals	$10\times10\times10$
CsI:Tl	Hilger crystals	$10\times10\times10$
CsI:Na	Hilger crystals	$10\times10\times10$
$CdWO_4$	Hilger crystals	$10\times10\times10$
$ZnWO_4$	Hilger crystals	$10\times10\times10$
$Lu_2(SiO_4)O$:Ce (LSO:Ce)	Hilger crystals	$10\times10\times10$
$Lu_xY_{2-x}(SiO_4)O$:Ce (LYSO:Ce)	Hilger crystals	$10\times10\times10$
Gd_2SiO_5:Ce (GSO:Ce)	Hitachi chemical	$10\times10\times10$
$Gd_xY_{2-x}(SiO_4)O$:Ce (GYSO:Ce)	Proteus, Inc.	$10\times10\times10$
$Y_3Al_5O_{12}$:Ce (YAG:Ce)	Hilger crystals	$10\times10\times10$
$YAlO_3$:Ce (YAP:Ce)	Hilger crystals	$10\times10\times10$
$LuAlO_3$:Ce (LuAP:Ce)	Hilger crystals	$10\times10\times10$
$Lu_3Al_5O_{12}$:Pr (LuAG:Pr)	Furukawa Co.	$10\times10\times10$
$LaBr_3$:Ce	ORNL*	*
$LaCl_3$:Ce	Saint-Gobain crystals	$10\times10\times10$
$CeCl_3$	ORNL*	*
$CeBr_3$	Schott AG	$10\times10\times10$
SrI_2:Eu^{2+}	ORNL*	*
BaF_2	Harshaw Chemical Co.	$10\times10\times10$

*Oak Ridge National Laboratory.
*Random shape with one flat polished face ($\sim$1 cm^3 volume).

It is important to emphasize that effectively all of the scintillators whose elevated temperature relative performance is studied here have previously been extensively and accurately characterized (particularly at or near room temperature) in terms of their pertinent gamma ray detection properties (e.g., light yield, energy resolution, emission wavelength(s), density, decay time, etc). These properties are well summarized with corresponding original reference citations in the excellent web-accessible scintillator compilation created and maintained by Stephen Derenzo, Martin Boswell, Marvin Weber, and Kathleen Brennan [4], and the reader is referred to this compilation and the references contained therein for data on the properties of