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Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeasDevelopment of BaF₂ transparent ceramics and evaluation of the scintillation propertiesTakumi Kato ^{a,*}, Go Okada ^a, Kentaro Fukuda ^b, Takayuki Yanagida ^a^a Graduate School of Materials Science, Nara Institute of Science and Technology (NAIST), 8916-5 Takayama-cho, Ikoma-shi, Nara 630-0192, Japan^b Tokuyama Corp., 1-1 Mikage-cho, Shunan-shi, Yamaguchi 745-8648, Japan

HIGHLIGHTS

- We have synthesized BaF₂ transparent ceramic by an SPS technique.
- We have characterized the optical, scintillation properties of BaF₂ transparent ceramic and single crystal.
- The scintillation light yield of the BaF₂ transparent ceramic was evaluated to be about 6000 photons/MeV under ¹³⁷Cs γ-rays.

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ABSTRACT

We have developed BaF₂ transparent ceramics by using a spark plasma sintering (SPS), and investigated optical, scintillation and dosimeter properties, in comparison with a BaF₂ single crystal. The photoluminescence emission peak of transparent ceramics prepared in a vacuum condition appeared around 500 nm. Under X-ray irradiation, emissions at 190 and 220 nm due to auger-free luminescence (AFL) were observed in the ceramic samples. The scintillation light yield of the BaF₂ transparent ceramics was evaluated to be about 6000 photons/MeV under ¹³⁷Cs γ-rays. In addition, the afterglow level of ceramic sample was lower than that of the single crystal. Thermally-stimulated luminescence glow peaks were observed at 50, 105, 140, 205, 280 and 420 °C in all the samples.

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1. Introduction

Inorganic scintillators, which convert high energy ionizing radiation to thousands of visible photons (Yanagida, 2013), have been playing a major role in many fields of radiation detection, including medical (Yanagida et al., 2010c), security (Totsuka et al., 2011), oil-logging (Yanagida et al., 2013b), environmental monitoring (Watanabe et al., 2015), astro- (Kokubun et al., 2004) and particle physics (Ito et al., 2007). In these applications, scintillators for γ-ray detections have attracted much attention especially for medical and security applications since these applications are very familiar to our daily life. Up to now, most scintillation detectors have consisted of single crystals mainly due to their high optical qualities. Except for Gd₂O₂S (Yoshida et al., 1988) used for X-ray CT and ZnS (McCloy et al., 2015) used for charged particle detectors, applications of ceramic materials are limited because the detection

efficiency is generally restricted by the opacity of ceramics. However, thanks to the advancement of the ceramic technologies, we now have a new way to deform crystalline powder into a bulk ceramic in a transparent form, which has a great advantage to be used as a scintillator.

Barium fluoride (BaF₂) crystal is known as a potential scintillator material. In the 1970s, Farukhi and Swinehart (1971) have first reported scintillation by BaF₂ crystal due to auger-free luminescence (AFL) and self-trapped excitation (STE), which appear around 190–220 and 310 nm, respectively. In particular, BaF₂ has considerably large effective atomic number ($Z_{\text{eff}} = 52.7$), so it has a great advantage to be used for γ-ray detections. However, compared with a conventional γ-ray scintillator (Lu_{2-x}Y_xSiO₅:Ce; 25,000 ph/MeV) used in PET, scintillation light yield of BaF₂ is much smaller (1500 ph/MeV). In addition, the emission spectral range of the AFL overlaps with that of STE, which has a much longer decay time constant (0.6 μs) (Blasse, 1994; Fedorov et al., 2016; Visser et al., 1993; Yanagida et al., 2010b). Most above studies have been performed using a single crystal form of BaF₂; and there are large room

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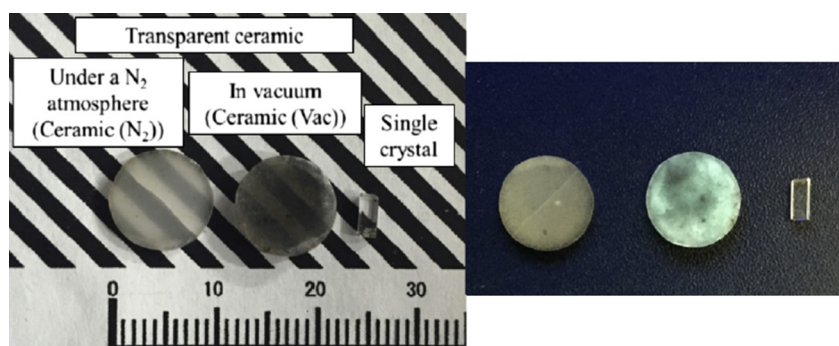


Fig. 1. BaF₂ ceramics and single crystal samples under room light (left) and UV (254 nm) light (right).

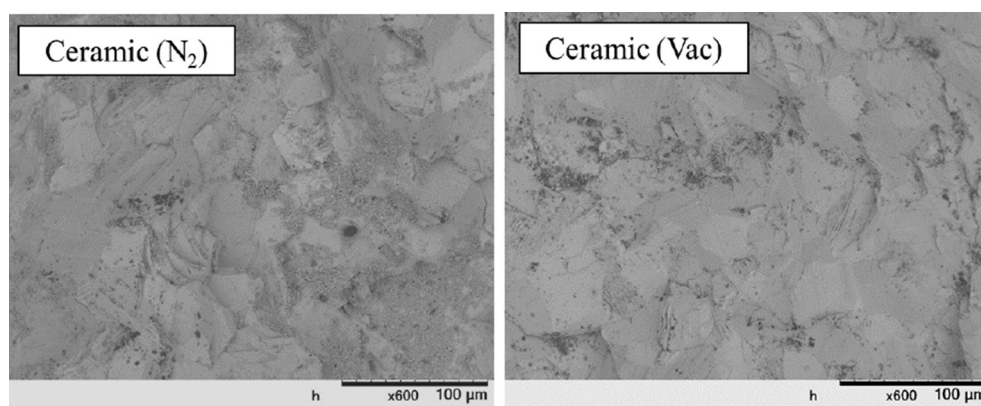


Fig. 2. SEM images of Ceramic (N₂) and (Vac).

for a development of BaF₂ transparent ceramics for scintillator applications. Earlier studies (Demidenko et al., 2010; Fedorov et al., 2016) have reported on non-doped and Ce-doped BaF₂ transparent ceramics; however, the scintillation in the vacuum-ultraviolet (VUV) range has not yet been evaluated and it has not been even known the scintillation in the VUV is present in a BaF₂ transparent ceramic, yet.

In this study, we developed the undoped BaF₂ transparent ceramics and characterized scintillation properties in relation with AFL. Specifically, the synthesis of BaF₂ transparent ceramics was carried out by the spark plasma sintering (SPS) method, and the optical and scintillation properties were characterized. Further, we have characterized the thermally stimulated luminescence (TSL) glow curve in order to study relatively shallow trapping centers. Up to now, our groups have succeeded to develop many kinds of transparent ceramic scintillators such as Ce:YAG (Yanagida et al., 2005), Ce:LuAG (Yanagida et al., 2011), Pr:LuAG (Yanagida et al., 2012), Ce:GAGG (Yanagida et al., 2013c) and Yb:Lu₂O₃ (Yanagida et al., 2013b). In these studies, most transparent ceramic samples showed superior scintillation properties than those of single crystals with the same chemical compositions; therefore, BaF₂ transparent ceramics are of great interest to investigate. In addition, to the best of our knowledge, no reports are found for bulk fluoride ceramics produced by SPS for scintillator applications.

2. Materials and methods

BaF₂ transparent ceramic samples were synthesized by the SPS method using Sinter Land LabX-100 as follows. First, BaF₂ powder (1.5 g) of a reagent grade was loaded in a graphite die and sealed with two graphite punches, and the graphite assembly was then

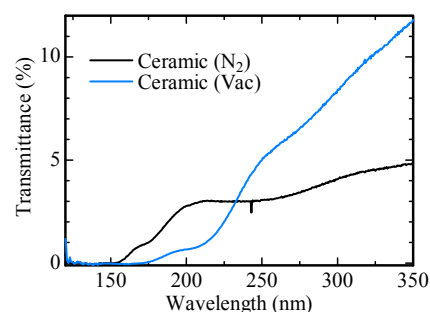


Fig. 3. Transmittance spectra of Ceramic (N₂) and Ceramic (Vac).

loaded to the furnace to sinter. The sintering temperature was controlled as described in the following sequence: the sintering temperature was increased from 20 °C to 720 °C at the rate of 24 °C/min and held for 30 min while applying the pressure of 10 MPa, and then the sintering temperature was increased to 960 °C at the rate of 24 °C/min and held for 45 min while applying the pressure of 100 MPa in a N₂ atmosphere or vacuum. In this paper, we denote the obtained samples Ceramic (N₂) and Ceramic (Vac) for the ones sintered in N₂ and vacuum, respectively. After the synthesis, the wide surfaces of the ceramic samples were polished. In the course of study, the following measurements were carried out for all the prepared samples, and some of the measurements were done for BaF₂ single crystal prepared by Tokuyama Corp. In these experiments, all the ceramics and the reference single crystals were synthesized by using the same BaF₂ powders.

Backscattered electron image of ceramic sample was obtained

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