

# Inorganic Scintillation Crystals

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inorganic scintillation crystal

radiation detector

temperature dependence

## 1. Introduction

Scintillators play a crucial role as radiation detection materials in various nuclear technologies and radiation applications, such as medical imaging, well logging, homeland security, marine and space exploration, and high energy physics (HEP). They indirectly detect radiation and are usually coupled with a photo-sensor. In a scintillator, the energy deposited by incoming radiation is converted into light photons, which are detected by a photo-sensor and converted into an electrical signal. Generally, scintillators can be classified into organic and inorganic, and the scintillator type used in a radiation detector is determined by the type of radiation particle to be measured as well as the purpose of radiation detection.

Organic scintillators, such as Stilbene and liquid scintillators, have an excellent pulse shape discrimination ability to distinguish between gamma rays and neutrons or alpha particles. Thus, they are mainly used to detect neutrons or accelerated charged particles, such as protons and alpha particles <sup>[1][2]</sup>. However, owing to the low density and detection efficiency (stopping power) of organic scintillators, inorganic ones are preferred when measuring X-rays or gamma rays. In terms of temperature, the melting points of inorganic scintillators are typically higher than those of organic scintillators, and most inorganic scintillators are grown in high-temperature furnaces. Because of their higher melting points, inorganic scintillators are more resistant to high temperatures than are organic scintillators <sup>[3]</sup>.

Inorganic scintillators are primarily ionic solids and composed of high-density crystals. They can be classified into two categories (single-crystals and polycrystalline ceramics), with the former typically exhibiting better optical properties at the expense of fabrication costs <sup>[4][5]</sup>. Polycrystalline ceramics' relatively poorer optical properties (transparency) often limit their applications to lower energy radiation detection where smaller-sized scintillators can be used.

Single-crystal inorganic scintillators are preferred in fields requiring radiation detection under extreme conditions (high radiation, temperature, humidity, vibrations, etc.), such as well logging, HEP, nuclear reactor monitoring, and

space exploration. In these applications, large-sized scintillators are often used to detect and measure high-energy radiation under harsh conditions.

In the well logging industry, the growing demand for fossil fuels worldwide has led to deeper drilling to search for new fuel sources, and the increasing depth of wells creates more challenging downhole environments. In addition, future HEP experiment environments are expected to be harsher in terms of radiation exposure. In this respect, development of new scintillators that can withstand higher temperatures and research on existing scintillators for use in extreme environments (high radiation, vibration conditions, humidity, etc.) are ongoing.

Therefore, we review and summarize single-crystal inorganic scintillator candidates, which can be used in several applications requiring radiation detection in extreme environments. Focus is placed on factors that directly affect scintillation properties (i.e., temperature dependence and radiation resistance) and physical properties such as susceptibility to mechanical shock (vibration) and hygroscopicity are also considered depending on application fields.

The temperature dependence of a scintillator is typically assessed by evaluating changes in the light yield of the scintillator with varying temperatures. Other general properties of the scintillator, including decay time and energy resolution, are also often considered. Similarly, the radiation resistance of a scintillator is assessed by the change in light yield or optical transmittance of the scintillator with respect to radiation dose. Therefore, radiation dose rate dependence of various scintillator candidates and radiation damage recovery via thermal annealing are also reviewed in this study. In addition, in applications where the detectors are exposed to high humidity, such as in a nuclear power plant (NPP) in the event of a severe accident, or high vibrations (such as well logging and space exploration), the hygroscopicity and mechanical susceptibility of the crystal (and subsequent components such as photosensor) should be considered.

## **2. Scintillation Crystal Applications in Extreme Environments**

### **2.1. Well-Logging Industry**

For decades, there has been a steady demand for high-temperature radiation detectors to be used in the well-logging industry, and oil wells need to be drilled deeper to access new fuel sources. The deeper the well, the harsher the downhole environment (temperature) in which nuclear measurements need to be made. Furthermore, in the case of logging while drilling implementations, the radiation detector may experience high levels of vibration and shock. In general, the downhole environment is known to be at a temperature of about 175 °C and a pressure of 20,000 psi, and the vibration level and shock level caused by drilling are ~30 g RMS and ~700 g, respectively [6]. Therefore, radiation detectors used in the well-logging industry must maintain their performance in terms of scintillation properties (such as emission spectrum, decay time, and light output) at high temperatures. In addition, radiation detectors have to operate at high levels of vibration and shock, so the brittleness of the scintillator should also be considered.

There have been numerous studies on the temperature dependence of single-crystal scintillators for the well-logging industry. NaI:TI—a traditional halide scintillator—is a commonly used scintillator in nuclear well-logging tools due to its high light output and good temperature dependence [7]. The temperature dependence of NaI:TI was evaluated up to 300 °C [8]; NaI:TI showed acceptable temperature dependence at temperatures between 175 °C and 200 °C. Although NaI(Tl) has been used in the well-logging industry for more than 60 yr because of its performance at high temperatures, high light yield, and low cost, it has a few critical drawbacks, such as low detection efficiency. Moreover, it requires thorough packaging due to its hygroscopicity and fragility [9].

The increasing desire for more efficient high-temperature-resistant scintillators has led to the discovery of new halide scintillators, such as LaBr<sub>3</sub>:Ce and LaCl<sub>3</sub>:Ce, which were first introduced in the early 2000s. These halide scintillators have received huge attention due to their excellent properties such as excellent light yield, good energy resolution, and high density. Regarding temperature resistance, LaBr<sub>3</sub>:Ce was reported to show 8% energy resolution at 175 °C, superior to the 9.9% energy resolution of NaI:TI at RT. Along with LaBr<sub>3</sub>:Ce, 10% doped LaCl<sub>3</sub> showed even more impressive scintillation characteristics over a wide range of temperatures. For example, LaCl<sub>3</sub>:Ce maintains an almost constant light yield from 100 to 600 K, reaching its maximum at 500 K [10]. Thus, these two scintillators can replace NaI:TI scintillator in well-logging, considering that they both have a similar drawback as NaI:TI of being extremely sensitive to humidity [11] and being very brittle [12].

Recently, the Cs<sub>2</sub>LiYCl<sub>6</sub>:Ce (CLYC) scintillator has drawn attention as a promising scintillator for well-logging because of its ability to detect both gamma rays and neutrons. At 120 °C, CLYC's light yield retention, relative to that at RT, is significantly better than that of NaI:TI. At 175 °C, CLYC maintained a value of 78% of that at RT, whereas NaI:TI was only 55% [13]. Moreover, CLYC's neutron detection capability maintains its decay time and light yield for neutron particles at temperatures up to 150 °C and only showed slight degradation at higher temperatures, making it a suitable scintillator for well-logging using neutron radiation [14].

Another major scintillator group that can be used in well-logging is oxide scintillators. Among oxide scintillators, the temperature dependence of the Ce:GSO scintillator was studied in 1991, and it demonstrated a high light output even at 175 °C [15]. In addition, Ce:YAP scintillator discovered in the 1980s [16] has many attractive properties, such as reasonably high density, fast decay, negligible afterglow, high light yield, and an excellent energy resolution of 4.4% for 662 keV gamma radiation. At 150 °C, the light output of Ce:YAP scintillator is half of that at RT. Generally, this scintillator shows a constant scintillation response with temperature change [17].

Some of the newly developed oxide scintillators have demonstrated potentials to be used in the well-logging industry. The Ce:GPS scintillator was first introduced in 2007 [18] and is known to have excellent energy resolution, light yield, and temperature dependence. The light yield of Ce:GPS was almost consistent from RT up to approximately 250 °C [19]. The light yield of Pr:LuAG, another scintillator known to have excellent temperature resistance, shows only slightly lower light output at 225 °C than at 50 °C. However, compared with Ce:GPS, the Pr:LuAG scintillator has a relatively low light yield and possesses intrinsic radioactivity and short peak emission wavelength (306 nm). Therefore, the Ce:GPS scintillator is expected to show more excellent performance than the Pr:LuAG scintillator in high-temperature environments, including well-logging. Another scintillator—Ce:LuAP—had

an observable photopeak at a temperature of 395 °C, and the performance at elevated temperatures indicated Ce:LuAP has potential in well-logging applications [20].

Vibration often accompanies well-logging, and vibration effects can be mitigated by selecting a robust scintillator or through better packaging of the radiation detector as a whole. Oxide based scintillators (such as Ce:YAP, YAG, etc.) are relatively less brittle than halide scintillators [21], but most halide scintillators, including lanthanum halide and elpasolite, are extremely brittle [12][22]. For example, NaI rates 2 on the Moh hardness scale, but LuAG(8.5), YAG (8.5), and YAP(8.6) are high on the Moh hardness scale [23]. Therefore, radiation detectors have to be properly packaged to withstand the vibration and the shock during geophysical oil logging operations. In particular, when using a halide based scintillator, it is necessary to improve the packaging by employing an internal shock resistant buffer. A list of suitable scintillators for the well-logging industry, along with their main characteristics, is presented in Table 1.

**Table 1.** Scintillator list and critical parameters for radiation detector used in the well-logging industry.

Crystal	Light Yield (Photons/MeV)	Density (g/cm <sup>3</sup> )	Relative Light Output <sup>1</sup>	Hygroscopicity	Ref
NaI:Tl	38,000	3.67	55% at 175 °C	Hygroscopic	[8]
LaBr <sub>3</sub> :Ce	65,000	5	90% at 175 °C	Hygroscopic	[11]
LaCl <sub>3</sub> :Ce	49,000	3.86	100% at 225 °C	Hygroscopic	[10]
CLYC	~20,000	3.31	78% at 175 °C	Hygroscopic	[8]
GSO:Ce	13,000	6.71	60% at 150 °C	Non-hygroscopic	[15]
Ce:YAP	~24,000	5.5	50% at 150 °C	Non-hygroscopic	[17][24]
Ce:GPS	30,000	5.5	100% at 250 °C	Non-hygroscopic	[19][18]
Pr:LuAG	~20,000	6.7	70% at 225 °C	Non-hygroscopic	[19][20]
Ce:LuAP	~4300	8.34	200% at 200 °C	Non-hygroscopic	[25]

2.2. HEP

In HEP, inorganic scintillators are essential for most radiation detectors in calorimeters, both existing and under development. The radiation involved in HEP is typically high-energy photons and particles in large numbers (high fluence rate). Therefore, characteristics required of detectors in calorimeters include high detection efficiency (density), fast decay time, good energy resolution, and radiation resistance for precise measurements of large numbers of high-energy radiation [26]. In HEP, scintillator light yield is secondary, as the energies involved are high, leading to a sufficiently large number of light photon emission during detection. Common inorganic scintillators that compose calorimeters used in HEP experiments include CsI (undoped), BGO, and PbWO<sub>4</sub> (PWO). Of these, the

Compact Muon Solenoid (CMS) particle detector (composed of 75,848 PWO scintillators) has a total size of 11 m<sup>3</sup>, and is the largest among the used calorimeters. With its excellent energy resolution (for the target high energy radiations) and detection efficiency (high density), the CMS PWO calorimeter played an essential role in discovering the Higgs boson via CMS experiments [27]. Future HEP experiment environments will be even higher radiation environment; thus, bright, dense, and fast scintillator detectors with excellent radiation hardness are required. As mentioned in Section 2.2, the light output of scintillators can decrease under high radiation conditions. As expected, a significant loss of light output from the PWO scintillator was observed in the CMS PWO calorimeter [28]. To operate in the High-Luminosity Large Hadron Collider (HL-LHC), a scintillator must survive an absorbed dose of 100 Mrad (100 Mrad), a hadron fluence of  $6 \times 10^{14} \text{ cm}^{-2}$ , and a fast neutron fluence of  $3 \times 10^{15} \text{ cm}^{-2}$  [29]. Numerous studies have investigated the degree of radiation damage on various inorganic scintillators between radiation doses ranging between 0 and 340 Mrad to find a scintillator that can survive at this level.

The light output of an undoped CsI decreased to 30% of its original value (decrease of 70%) after a gamma-ray irradiation dose of 1 Mrad but showed only an approximately 20% light output drop after a dose of 100 krad, indicating that the undoped CsI had radiation hardness against gamma-ray irradiation up to a 100 krad dose [30][31]. Since the radiation damage of the undoped CsI was not recovered at RT, it was dose-rate independent. Therefore, it is possible to calibrate an undoped CsI calorimeter using light monitoring. In addition, undoped CsI has a fast decay time of approximately 30 ns and is suitable for mass production because of its low manufacturing cost. These advantages made undoped CsI to be selected as the scintillator in the calorimeter of Fermilab's KTeV experiment [32].

Moreover, the radiation damage of BGO [33] and PWO [34] recovers at RT after several hours or weeks, so they are dose-rate dependent. The light output of BGO and PWO scintillators, respectively, decreased to 45% and 30% of their original values at a 120-Mrad dose.

Studies regarding proton and neutron irradiation on PWO scintillators have also been conducted [35]. In this study, the PWO scintillator had an induced absorption length of  $\sim 15 \text{ m}^{-1}$  after proton irradiation with a fluence of  $5 \times 10^{13} \text{ cm}^{-2}$ , whereas the induced absorption length of PWO was  $0.3 \text{ m}^{-1}$  at a gamma-ray dose of 5 Mrad, which showed that PWO exhibited less radiation hardness for protons compared with gamma rays. In recent neutron irradiation experiment of PWO, approximately 86% of the light output loss was observed in PWO after  $1.6 \times 10^{15}$  fast neutrons/cm<sup>2</sup> irradiation [36]. In the same study, the LYSO scintillator showed significantly higher radiation hardness for neutrons compared with the PWO scintillator, with less than 25% light loss observed even after irradiation of up to  $9 \times 10^{15}$  fast neutrons/cm<sup>2</sup>. LYSO maintained 75% light output even after 120 Mrad of gamma-ray irradiation, and the radiation damage of LYSO was dose-rate independent [31]. Following these results, LYSO crystals were proposed as the scintillation materials for an LYSO/W Shashlik sampling calorimeter in the CMS upgrade for the HL-LHC [37], and total-absorption LYSO crystal calorimeters were proposed for the SuperB experiment in Europe [38] and Mu2e experiment at Fermilab [39].

The radiation resistance of other oxide inorganic scintillators, such as Ce:GPS, GSO, and Pr:LuAG, has also been investigated. Ce:GSO scintillators have excellent radiation resistance and fast decay time. It was reported that

Ce:GSO did not show a noticeable decrease in light yield up to 100 Mrad [40], but another study reported an increase in the light output of Ce:GSO after gamma-ray irradiation [41]. The Ce:GPS scintillator was reported to show 57% and 15% light output of their original values after gamma-ray irradiation of approximately 68 and 369 Mrad, respectively; in addition, Pr:LuAG scintillator showed 46% and 36% light output at gamma-ray irradiation of approximately 70 and 382 Mrad, respectively [19]. According to [1], in the order of increasing radiation resistance, are thallium-activated alkali halides, CsF, BGO, YAO, CeF<sub>3</sub>, BaF<sub>2</sub>, and GSO. Table 2 summarizes the essential properties of selected scintillators and their radiation hardness regarding HEP experiments.

**Table 2.** Performances of selected scintillators and critical parameters for HEP experiments.

Crystal	Decay Time (ns)	Density (g/cm <sup>3</sup> )	Relative Light Output <sup>1</sup> at Radiation Dose (%)	Dose-Rate Dependence	Ref
LYSO	40	7.4	89% at 1 Mrad 75% at 120 Mrad	X	[31] [42]
Pr:LuAG	20	6.7	46% at 70 Mrad	O	[19] [43]
Ce:GPS	46	5.5	57% at 68 Mrad	X	[19]
GSO	30	6.7	100% at 100 Mrad (No degradation)	O	[40] [41]
PWO	30 6	8.3	30% at 120 Mrad	O	[34]
Undoped-CsI	30	4.5	80% at 100 krad 30% at 1 Mrad	X	[30]
BGO	300	7.1	45% at 120 Mrad	O	[33]
BaF <sub>2</sub>	650	4.9	40% at 120 Mrad	X	[44]

<sup>1</sup> Compared with the radiation undamaged scintillator.

As future HEP experiment environments will be harsher in terms of radiation exposure, it is expected that fast, dense, bright, and radiation-resistant scintillators will continue to play a crucial role in HEP experiments. Therefore, related R&D of radiation-resistant scintillators is expected to continue.

2.3. Nuclear Reactor Monitoring System in Nuclear Power Plant

Nuclear power plants (NPPs) have been constructed globally to meet the ever-increasing demand for energy. As of 2020, 442 NPPs were operating in 30 countries, and the commissioning of 52 new NPPs in 15 countries is underway [45]. As the number of NPPs in operation and under construction increases, there is an increasing interest in the safety of these plants. In particular, after the Three Mile Island accidents in 1979 and the Chernobyl

disaster in 1986, accident management has been crucial for NPPs. After these accidents, an NPP accident monitoring system had been designed and installed using guidelines that included the impact of lessons learned from the accidents. Despite this, another severe accident at the Fukushima Daiichi NPP in March 2011 resulted in many severe failures, such as power outages in several monitoring devices, reactor core damage, and hydrogen explosions. Therefore, it was necessary to review the standards of equipment used for NPP accident monitoring. Accordingly, IAEA established an action plan for nuclear safety in response to the Fukushima Daiichi accident and provided instructions for severe accident monitoring systems in NPPs [46].

These guidelines focused on maintaining the integrity of the reactor core, reactor pressure vessel (RPV), and reactor containment vessel (CV) conditions due to the experience acquired from the damaged Fukushima Daiichi reactor CV. In this guideline, the severe accident plant state for boiling water reactor (BWR) and pressurized water reactor (PWR) plants were classified into four severe accident states (SAs). The definitions and environmental conditions of each state suggested in the guideline are shown in Table 3 [47].

Table 3. Severe accident states—SA1 to SA3b—for NPP reactors.

Reactor Type and Location		SA1 <sup>1</sup>	SA2	SA3	
				SA3a	SA3b
BWRs	Plant condition	Core damage (Meltdown)	Core damage RPV damage	Core damage	Core damage
				RPV damage	RPV damage
				PCV damage	PCV damage
	Condition <sup>2</sup> in PCV <sub>3</sub>	171 °C	300 °C	700 °C	1000 °C
		500 Mrad/6 month	500 Mrad/6 month	500 Mrad/6 month	500 Mrad/6 month
		Steam	Steam	Steam	Steam
	Condition outside PCV	66 °C	66 °C	100 °C	100 °C
		30 Mrad/6 month	30 Mrad/6 month	200 Mrad/6 month	200 Mrad/6 month
		100%	Steam	Steam	Steam
	PWRs	Plant condition	Core damage RV damage	Core damage	Core damage
				RV damage	RV damage
				CV damage	CV damage
		Condition in CV <sup>4</sup>	190 °C	200 °C	200 °C
		-	200 Mrad/yr	200 Mrad/yr	200 Mrad/yr



Reactor Type and Location	SA1 <sup>1</sup>		SA2		SA3	
					SA3a	SA3b
	100%		100%		100%	100%
	Condition outside CV	Atmospheric condition	-		-	-

<sup>1</sup> SA—severe accident. <sup>2</sup> Environmental condition—Maximum temperature, radiation dose, and humidity, respectively. <sup>3</sup> PCV—primary containment vessel. <sup>4</sup> CV—containment vessel.

SA3 (RPV injury) was divided into two states. One is SA3a, which included the type of accident that occurred at the Fukushima Daiichi NPP, and the other is SA3b, which was considered beyond SA3a. SA3a and SA3b were differentiated based on the success of the accident management strategy (early water injection within 24 hr after core damage). These severe state stages were intended to identify the criteria for designing accident monitoring devices necessary to facilitate the mitigation of accident progression. Therefore, equipment capable of monitoring the reactor must operate under high temperature and radiation conditions specified in Table 3.

Because of the extreme environment around reactors, the radiation monitoring system of NPPs usually monitors the radiation level in specific areas (area radiation monitoring system) or radioactive fluid and effluent in the plant (process radiation monitoring system) outside the containment rather than monitoring the reactor within the containment. In this section, we present potential scintillators that could be used as radiation detection materials for nuclear reactor monitoring within the containment of PWR.

As described above, unlike in fields such as HEP and well-logging, scintillators used in reactor monitoring systems must be resistant to both high temperature and radiation. In addition, since radiation damage of a scintillator can be recovered at high temperatures, one should also consider scintillator radiation damage recovery via thermal annealing. On the other hand, during a nuclear accident severe accident, the humidity in the NPP containment building is expected to be very high (100% or steam) and the hygroscopicity of scintillator should also be considered (Table 3 and Table 4). Furthermore, according to the guideline by IAEA [\[48\]](#), radiation detectors for use during a severe accident are expected to be grouped in “seismic category 1”, which means that they should be designed to withstand vibrations as defined by “seismic level 2”—the most stringent seismic safety requirements for a NPP.

**Table 4.** Performances of selected scintillators and critical parameters for a nuclear reactor monitoring system.

Crystal	Light Yield (Photons/MeV)	Relative Light Output <sup>1</sup> at Radiation Dose (%)	Maximum Temperature	Thermal Annealing Effect	Hygroscopicity	Ref
LYSO	33,200	73% at 383 Mrad	150 °C	Full recovery (above 400 °C)	Non-hygroscopic	<a href="#">[19]</a> <a href="#">[31]</a> <a href="#">[42]</a>



Crystal	Light Yield (Photons/MeV)	Relative Light Output <sup>1</sup> at Radiation Dose (%)	Maximum Temperature	Thermal Annealing Effect	Hygroscopicity	Ref
Pr:LuAG	24,000	36% at 382 Mrad	225 °C	Partial recovery (above 400 °C)	Non- hygroscopic	<a href="#">[19]</a> <a href="#">[49]</a>
<sup>1</sup> Ce:GPS	30,000	15% at 369 Mrad	350 °C	Full recovery (above 400 °C)	Non- hygroscopic	<a href="#">[19]</a> <a href="#">[18]</a> <a href="#">[50]</a>

A study reported the temperature dependence and radiation resistance of several inorganic scintillators under severe NPP accident conditions (Table 3). In the study, the temperature dependence, radiation resistance, and radiation damage recovery via thermal annealing were investigated for Pr:LuAG, LYSO, and Ce:GPS scintillators [\[19\]](#), and all of these three scintillators are non-hygroscopic. The Ce:GPS scintillator showed about 15% light output of its original value (decrease of 85%) after 369-Mrad gamma-ray irradiation; the Pr:LuAG and LYSO scintillators showed light outputs of 36% and 73%, respectively, after 380-Mrad gamma-ray irradiation. Therefore, of these scintillators, LYSO exhibited the strongest radiation resistance, and Ce:GPS exhibited the weakest radiation resistance. Moreover, in the temperature dependence evaluation of these scintillators, the maximum temperature at which the photopeak of a Cs-137 radiation source was observed (maximum observable photopeak temperature) for the Ce:GPS scintillator was 350 °C, much higher than those of the Pr:LuAG and LYSO scintillators (225 °C and 150 °C, respectively). Particularly, Ce:GPS demonstrated almost consistent light output from RT up to approximately 250 °C, and it showed rapid radiation damage recovery with more than 300 °C thermal annealing. These properties of Ce:GPS revealed its potential to be employed under SA2 conditions (300 °C, 500 Mrad/6 month) in a BWR NPP PCV, and SA3b conditions (300 °C, 200 Mrad/year) in a PWR NPP CV.

2.4. Space Exploration

Gamma-ray spectroscopy (GRS) has been used in space exploration to study the composition of the surface of airless solar system bodies, such as the Moon, Mars, Mercury, and large S-class asteroids [\[51\]](#)[\[52\]](#)[\[53\]](#). To perform GRS in space exploration, the gamma-ray detector should meet the criteria in terms of detection efficiency, energy resolution, and reliability. Since gamma-ray detectors used in space exploration are often exposed to high radiation environments arising from sources, such as galactic cosmic rays and solar flares, the radiation resistance of scintillators is crucial to ensure their reliability.

For example, the BepiColombo mission was a joint mission held by the European Space Agency and Japan Aerospace Exploration Agency to perform remote GRS of Mercury’s surface and determine the elemental composition of the planet [\[54\]](#). According to a study conducted to search for alternatives to traditional scintillators for space GRS [\[55\]](#), an ideal detector should possess the following properties—8-cm minimum gamma-ray pathlength, >5-g/cm<sup>3</sup> high density, excellent energy resolution of ≤3% for 662 keV gamma radiation, and peak detection efficiency of >6% at 6 MeV. In addition, it has to be proton radiation resistant to the 100-krad level.

Traditional scintillators, such as NaI:TI and CsI:TI, had insufficient energy resolution for the accurate distinction of the formation ions. Therefore, the demand for higher energy resolution, light yield, and radiation-resistant properties motivated researchers to search for alternative scintillators. A study that investigated the LaBr<sub>3</sub>:Ce scintillator for the BepiColombo mission found that LaBr<sub>3</sub> doped with 5% cerium concentration (LaBr<sub>3</sub>:5%Ce) showed stable performance in its light yield and energy resolution against a high proton radiation environment (100 MeV with a fluence of 10<sup>12</sup> protons/cm<sup>2</sup>) [56][57][58]. However, despite its excellent energy resolution and proton radiation-resistant properties, LaBr<sub>3</sub>:Ce had the drawback of being intrinsically radioactive (~1 Bq cm<sup>-3</sup>) due to the presence of <sup>138</sup>La [58]. To reduce this background noise, the Ce-doping concentration was increased until it completely replaced the lanthanum atom in LaBr<sub>3</sub>:Ce to yield CeBr<sub>3</sub>, which mitigated the internal activity of LaBr<sub>3</sub>:Ce by around a factor of 30 at the cost of reduced energy resolution. Up to the energy level of 3 MeV, CeBr<sub>3</sub> had better minimum detection limits than LaBr<sub>3</sub>:Ce, and both scintillators proved to have much greater detection limits than high purity germanium semiconductor detectors. In addition, CeBr<sub>3</sub> showed degradation from a gamma dose of 100 krad and was more gamma-ray radiation-resistant than LaBr<sub>3</sub>:5%Ce [59] but not significantly different in proton radiation hardness [60]. With respect to thermal dependence, LaBr<sub>3</sub>:Ce was shown to be more stable than that of CeBr<sub>3</sub> [61].

The gamma large array space telescope (GLAST) calorimeter, operated at low earth orbit (600 km above the surface of the Earth) can provide information on the energy of electromagnetic showers through pair conversion reactions from gamma rays interaction in the tracker. Therefore, the calorimeter can measure energy and provide directional information for gamma rays ranging from 10 MeV–300 GeV [62]. Thus, scintillators should measure the wide range of energy; they should also be cost-effective and easier to grow in large sizes or long lengths. In addition, the scintillators should be resistant to the radiation environment, especially protons. At that altitude, LaBr<sub>3</sub>:Ce could also be used for LEO missions [63][64]. Proton doses accumulated up to 5 years did not cause huge radiation damage to LaBr<sub>3</sub> and LaCl<sub>3</sub>, with an acceptable amount of activation [64][65].

CsI:TI for the Fermi Gamma-ray Space Telescope calorimeter was reported to be one of the most common scintillators for calorimeters in space [66]. In a study on radiation resistance of CsI:TI, CsI:TI crystal's light yield tended to decrease rapidly to the first 20 Gy level. Specifically, tests with gamma rays and protons recorded (24 ± 4)% and (22 ± 5)% light yield decreases at 180 and 175 Gy doses, respectively. These records passed the quality assurance tests to be used for space calorimeters by having the dose of 10 and 10<sup>4</sup> times higher than the ones seen in the orbit environment. Notably, the damages or displacements of the crystals due to gamma-ray irradiation could be partly recovered via thermal annealing, but not for proton irradiation [67]. In addition, 96 crystals of CsI were tested for their thermal stabilities showing no degradations in performance between -30 °C to 50 °C, and the mechanical stabilities (primary fundamental mechanical frequency of ~180 Hz) surpassed the 100 Hz vibration requirement that occurs during launch [68]. CsI were treated with wrappings around crystal to withstand the different expansion due to different thermal coefficients, and a series of mechanical tests were conducted to qualify for the mission environments [69].

The Dark Matter Particle Explorer (DAMPE) experiment was launched in 2015, while the High Energy Cosmic Radiation Detection (HERD) experiment is planned to be installed on the Chinese Space Station. Utilizing the

CALOCUBE electromagnetic calorimeter [70], DAMPE detects electrons and photons in the 5 GeV–10 TeV energy range for clues regarding dark matter and the origin of high energy cosmic rays [71][72], with an energy resolution of 1.5% at 800 GeV in space [73]. For this mission, they use the crisscross structure that consists of long plastic scintillator logs with two photomultiplier tubes attached to the ends. In addition, a BGO calorimeter suppresses back-splash fake events [74]. The plastic scintillator efficiently measures the particle charge and discriminate photons and electrons while BGO is utilized for the discrimination between electrons and protons from the electron and hadron showers with the help of neutron detectors rejecting protons in background.

The DAMPE satellite, in orbit for several years during the mission, is designed to be resistant against a total dose of 20 krad [75] and exposure to temperature ranges of −20 °C to +45 °C when in storage and −10 °C to +30 °C when in operation [74]. For use in the mission, radiation detector modules have been first put through the modal analysis to evaluate its resistance against deformations and stress, and recorded 128.4 Hz more than the required first order modal frequency of 70 Hz. In addition, to withstand the vibrational conditions during the mission, at least 1.24 mm, 6 g max (sweeping speed of 4 oct/min of 5–8 Hz and 8–100 Hz), 0.05 g<sup>2</sup>/Hz and 6.41 Grms (Duration 1 min, 20–100 Hz, 100–600 Hz, and 600–2000 Hz) were needed for the sinusoidal and random tests according to the acceptable level criteria [74].

With regards to radiation hardness, BGO’s afterglow increased only by around 7% up to 100 krad dose, compared to the 9200% increase in afterglow for GAGG:Ce [76]. The BGO scintillator responds to energies ranging from 10 MeV to 2 TeV and reported a temperature dependent light output change of −1.2% per degree Celsius around 0 °C [75], and −2.2% per degree Celsius in the ATIC experiment [77]. Because of this temperature dependence, four faces of the satellite are protected by thermal insulation foils and orbits synchronously with a single radiating surface to mitigate temperature fluctuations [74]. A temperature variation of 50 °C in space causes 4 mm change in the detector modules’ lengths due to the difference in thermal coefficients of honeycombs as protectors and scintillators. Therefore, special chips in the middle and the U-shape clamp are applied to reduce the frictions.

Recently, a relatively new scintillator, GAGG:Ce [78], has been reported to be a potential candidate for the LEO mission. Because of the high density (6.63 g/cm<sup>3</sup>), non-hygroscopicity, high light yield (56,000 photon/MeV), and good energy resolution—all of which are superior to those of CsI:TI—applications of GAGG:Ce have been investigated. However, the drawback of high afterglow after long exposure to proton environments has been observed. To mitigate this proton activation phenomenon, Mg co-doping has been employed [76]. A list of suitable scintillators for space applications, along with their main characteristics with focus on radiation tolerances, is presented in Table 5.

**Table 5.** Performances of reported scintillators and critical parameters for space exploration.

Crystal	Decay Time (ns)	Density (g/cm <sup>3</sup> )	Relative Light Output <sup>1</sup> at Radiation Dose (%) for Proton	Relative Light Output <sup>1</sup> at Radiation Dose (%) for Gamma-Ray	Ref
CsI:TI	680	4.51	78% at 18 krad	30–80% at 100 krad	[67]

Crystal	Decay Time (ns)	Density (g/cm <sup>3</sup> )	Relative Light Output <sup>1</sup> at Radiation Dose (%) for Proton	Relative Light Output <sup>1</sup> at Radiation Dose (%) for Gamma-Ray	Ref
					[76]
GAGG:Ce	100	6.63	88% at 100 krad	90% at 100 krad	[76] [79]
LaBr <sub>3</sub> :Ce	15.0	5.07	100% at 1 Mrad(No degradation)	92% at 100 krad	[80] [57] [81]
CeBr <sub>3</sub>	18.7	5.18	100% at 1 Mrad(No degradation)	98.6% at 100 krad	[59] [81]
BGO	300	7.1	~80% at 1.2 Mrad	65–90% at 100 krad	[82] [83]

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<sup>1</sup> Compared with the radiation undamaged scintillator

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