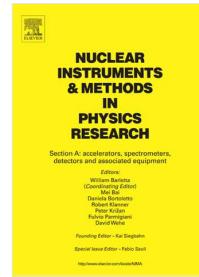


Journal Pre-proof

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1 Light yield, long-term stability, and attenuation length of a new
 2 plastic scintillator cured at room temperature

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15

16 **Abstract**

17

18 We, in collaboration with Carlit Holdings Co., Ltd., developed a new plastic scintillator that can be
 19 cured at room temperature by mixing a base resin with a hardener solution. We evaluated the
 20 scintillator's characteristics of light yield, stability, and attenuation length using a ⁹⁰Sr β-ray source.
 21 The plastic scintillator showed 40% of the light yield of the commercially available scintillator BC408,
 22 or 26% of that of the standard organic scintillator anthracene. The long-term light stabilities of the
 23 plastic scintillators, with and without the doping stabilizer agent 2,6-di-*tert*-butyl-4-methylphenol
 24 (BHT), were measured for 6 months. The BHT-doped sample retained 80% of the initial light yield
 25 over this period. The initial attenuation length of 0.9 m was obtained. The new plastic scintillator
 26 showed good stability against oxidative environments, although improvements in its light yield and
 27 attenuation length could be achieved by further optimizing its component materials and processing.

28

29 **Keywords:** Plastic scintillator, Room-temperature curing resin, Light yield, Long-term stability,
 30 Attenuation length

31

32 **1. Introduction**

33

34 Scintillation counters have been used as radiation detectors in high-energy and nuclear physics for
 35 many years. They have recently been applied as beam monitors in medical treatment [1–3], nuclear
 36 reactors [4, 5], and environmental radiation monitors [6]. Solid plastic scintillators are more easily
 37 handled and processed into various forms than liquid scintillators; however, they are relatively
 38 expensive. As plastic scintillator technology has matured, studies have been conducted to reduce
 39 manufacturing cost [7], improve radiation tolerance [8], and enhance neutron sensitivity [9–11] in
 40 recent years. In addition, lower-cost plastic scintillators are preferred for developing large-size
 41 calorimeters in high-energy physics [12–14] and neutrino detectors for reactor monitoring
 42 experiments [15–17].

43 In several projects, gadolinium-doped liquid scintillators have been adopted for increased neutron
 44 sensitivity from their large cross-sections by achieving large volumes [18–20]. However, because
 45 liquid scintillators are composed of flammable oil, avoiding the placement of large volumes of liquid
 46 scintillators near nuclear power plants is preferred. From this perspective, we developed the lower-
 47 cost gadolinium-doped plastic scintillator to facilitate a large volume of approximately one cubic
 48 meter [21].

49 In this study, we measured the light yield, attenuation length, and long-term stability of the newly
 50 developed plastic scintillator cured at room temperature and compared these properties with those of
 51 the commercially available standard BC408 (Saint-Gobain Co.) plastic scintillator.

52 The new plastic scintillator is advantageous because room-temperature curing avoids the high-

53 temperature processes required for standard commercial plastic scintillators, thus decreasing
 54 manufacturing costs and simplifying the facility requirements. In addition, freedom in shaping is
 55 increased because a broader variety of mold materials can be used. Furthermore, heat-labile functional
 56 materials can be added to improve the specific particle detection efficiency of the plastic scintillator.
 57

58 **2. Materials and methods**

59 **2.1. New manufacturing method**

60 Standard commercial plastic scintillators are usually composed of polystyrene or polyvinyl toluene
 61 with wavelength shifters. 2,5-Diphenyloxazole (PPO) and 1,4-bis(2-methylstyryl)benzene (bis-MSB)
 62 are used as the primary and secondary wavelength shifters, respectively. When plastic scintillators are
 63 made by casting or extrusion methods, these materials are heated to about 100°C or more [10]. After
 64 heating, plastic scintillators are cooled slowly to prevent cracking.
 65

66 We have developed a new plastic scintillator in collaboration with Carlit Holdings Co., Ltd., with a
 67 focus on reducing the production cost. The new plastic scintillator uses a bisphenol A-type epoxy
 68 resin as the base material, which can be cured at room temperature by mixing with an amine-based
 69 curing agent containing wavelength shifters (PPO, bis-MSB) and an antioxidant agent. The density
 70 and refractive index of the new plastic scintillator are $(1.15\text{--}1.16)\pm0.02\text{ g/cm}^3$ and 1.60 ± 0.04 (for
 71 450-nm laser light), respectively, almost equal to those of the standard BC408 scintillator, which has
 72 the density and refractive index of 1.032 g/cm^3 and 1.58, respectively [22].
 73

74 In this study, we used different scintillator base materials and modified the manufacturing
 75 procedure from that used in a previous study [21] to improve the transparency and reduce the
 76 variation in the samples. In addition, we added the antioxidant agent of 2,6-di-*tert*-butyl-4-
 77 methylphenol (BHT) and performed a post-curing process of holding for 6 h at 58 °C after room-
 78 temperature curing in order to improve the stability of light yield and attenuation length. We
 79 measured the light yield and the stability thereof of the new scintillator samples compared to those of
 80 BC408. All tested new plastic scintillators were fabricated using base materials from the same
 81 manufacturing lots.

82 **2.2. Experimental setup**

83 The experimental setup for measuring the light yield is shown in Fig. 1. The plastic scintillator
 84 samples prepared for this study are summarized in Table 1. We prepared six types of $40\text{ mm}\times75\text{ mm}$
 85 $\times3\text{ mm}$ plastic scintillator samples for light yield measurement (Fig. 2). Each plastic scintillator
 86 sample was shaped with a silicone rubber mold to improve the shape accuracy and reduce cutting
 87 uncertainty. The silicone frame comprised a 3-mm-thick silicone rubber sheet that was die-cut to 75
 88 $\text{mm}\times40\text{ mm}$. In addition, we applied an air bubble removal process for the sample scintillators after
 89 mixing the resin and hardener.
 90

91 We prepared two scintillator samples for each type. The sample with better air bubble removal was
 92 named “-1” and the other was named “-2”. As a reference, BC408 scintillators were also prepared
 93 with the same shape. Four sides of each sample scintillator plate were polished by an abrasive film
 94 and the $40\text{ mm}\times3\text{ mm}$ side was connected to a photomultiplier tube (PMT) using optical grease
 95 (BC630, Saint-Gobain Co.). During the measurement, each plastic scintillator was covered with an
 96 aluminized mylar sheet and a black sheet to increase the light yield and shield the scintillator from
 97 external light, respectively.

98 To select β-rays emitted from a ^{90}Sr β-ray source ($E_{\max}=2.28\text{ MeV}$, 1.7 MBq) that penetrated a 2-
 99 mm-diameter aluminum collimator of 10 mm in thickness and the sample scintillator, a trigger
 100 scintillator (BC408, $40\text{ mm}\times40\text{ mm}\times2\text{ mm}$) was set under the sample scintillator (Fig. 1). The
 101 PMTs of the sample and trigger scintillation counters were closely confined in a black box.
 102

103 In the experiment, the β-ray loses energy in the sample scintillator, releasing scintillation light. The
 104 scintillation light propagates through the scintillator and enters into a photocathode of the PMT
 105 (H7195, Hamamatsu Photonics K.K.) that is supplied with -2100 V . A signal charge from the PMT is
 106 converted to a digital signal by a charge-sensitive analog-to-digital converter (CSADC, V005, 14-bit,
 107 HOSHIN ELECTRONICS CO., LTD.). The ADC has the dynamic range 0–1000 pC and receives

108 gate signals of 200 ns derived from the trigger scintillation counter. The irradiation position of the β -
 109 ray is located 6 cm from the PMT surface on the centerline in the length direction of the scintillator.
 110 Each measurement was 5 min in duration.

111 One of the most important objectives of this study is the investigation of the long-term stability of
 112 the light yield of the new plastic scintillators. Scintillator samples were placed in a shading bag when
 113 not being tested and stored in a desiccator with the average temperature of 19.2 °C. Seasonal
 114 variations of the temperature spanned 10.4 to 24.3 °C.

115 A motorized stage (position accuracy of $\pm 15 \mu\text{m}$, OSMS26-200(X), SIGMAKOKI CO., LTD.) was
 116 used for highly precise positioning of the β -ray source and trigger scintillation counter (Fig. 1).

117 3. Results

120 3.1. Stability of light yield

122 The pulse shapes of the sample plastic scintillators, as observed by an oscilloscope, are shown in
 123 Fig. 3. One scintillator is selected from the two samples of each type. The light signal of this
 124 scintillator under β -rays is observed and the average pulse shape is recorded.

125 As shown in Fig. 3, the pulse widths of the new plastic scintillators are similar to that of BC408 at
 126 approximately 10–20 ns. The light yield is evaluated as the signal pulse height (PH). The PH is
 127 calculated as the signal-mean subtracted from the pedestal-mean (Fig. 4). The seven graphs in Fig. 4
 128 are acquired using samples with names ending in “-1”.

129 The mean values found from the histograms and the ratios of these mean values to the average
 130 mean value of BC408 are shown in Table 2. The mean values (ch/mm) are normalized by the sample
 131 scintillator thickness measured at the β -ray irradiation position using a micrometer to within an
 132 accuracy of 0.002 mm. The light yields of the new plastic scintillators are defined as the mean values
 133 (ch/mm) and are 33–46% of those for BC408. The light yields measured here are approximately two
 134 times higher than that in a previous study [21].

135 The long-term stability of the light yield is measured with results as shown in Fig. 5. The values of
 136 all data points are normalized by the initial value to evaluate the stability as a percentage. The data are
 137 plotted using two symbols to distinguish the two scintillator samples of each type. In Fig. 5, each
 138 sample pair was measured on almost same date and time. The error in the light yield measurements is
 139 estimated as the standard deviation of the results of the thirty measurements of BC408-1. The relative
 140 error is 2.6%.

141 As shown in Fig. 5, the light yield is generally stable for antioxidant-containing new scintillators.
 142 The influence of environmental oxidation is suppressed by the antioxidant. The light yield of the
 143 antioxidant-added new plastic scintillators maintains 80% of the initial value over at least 6 months.
 144 This indicates that the antioxidant stabilizes the new plastic scintillator.

145 3.2. Stability of attenuation length

148 The attenuation length of the scintillation light was measured [21, 23, 24]. The sample scintillator
 149 size was 20 mm \times 200 mm \times 3 mm, as in our previous study [21]. The most stable new plastic
 150 scintillator type, as shown in Fig. 5, is NGT6; therefore, the attenuation length of the NGT6L
 151 scintillator was measured and compared to that of the standard BC408L, which was cut from the
 152 BC408 scintillator plate. We added “-1” to the sample name of the one with fewer air bubbles and “-2”
 153 to the other. Four sides of each sample scintillator were polished using a grinder and one of the 20 mm \times
 154 3 mm sides was connected to a PMT using optical grease (BC630, Saint-Gobain Co.), as with the light
 155 yield measurement specimens in Table 1.

156 The method for measuring attenuation length was similar to that used previously [21], with the same
 157 setup as shown in Fig. 1. The irradiation position of the β -ray was 6 to 18 cm from the PMT surface.
 158 The detected light yield is defined as the mean value and normalized by the thickness at the irradiation
 159 position of the β -ray (Table 3). The scintillator samples were stored as described in section 2.2. The
 160 measurement results are shown in Fig. 6. The mean value of the light yield, which depends on
 161 the irradiation position, is fitted with equation (1):

$$f = \left(A + \frac{B}{x^2} \right) \cdot e^{-x/L}, \quad (1)$$

162 where A and B are amplitudes, x is the horizontal distance between the irradiation position and the
 163 PMT surface, and L is the light attenuation length [21]. In Fig. 6, the error in the light yield
 164 measurements is estimated as the standard deviation of five measured values from BC408L, as in the
 165 previous study [21]. The relative error is 1.3%. The standard deviation of the light yield in each data
 166 point is independent of the distance between the PMT surface and β -ray incident position and equal to
 167 1.3%; this is attributed to errors in each data point in the attenuation length measurement. For the
 168 sample scintillators NGT6L-1,2, the first measurements are performed approximately one month after
 169 the demolding of the scintillators, while the last measurements are obtained about eight months later.
 170 For BC408L, the last measurement is performed one year after the first measurement.

171 The storage times of the sample scintillators at measurement and the results of the attenuation
 172 length measurements are shown in Table 4 and Fig. 7. In Fig. 7, the BC408L measurement day differs
 173 from that of NGT6L-1 and -2. NGT6L-1 is manufactured with a high-quality polishing process and
 174 has fewer air bubbles than NGT6L-2. The long-term stability of the attenuation length could be
 175 improved, although the NGT6L-1 scintillator showed the initial attenuation length of approximately
 176 0.9 m. The specified attenuation length of BC408 is 2.1 m [22]. However, the BC408 sample used for
 177 the specification is large (1 cm \times 20 cm \times 200 cm), we measure a different initial attenuation length of
 178 0.94 m.

179 4. Summary

180 We developed a new plastic scintillator using room-temperature-curable resin. The light yield of
 181 the new plastic scintillator was approximately 40% of that of the standard commercial BC408 and
 182 26% of that of anthracene.

183 The light yield of the new plastic scintillator containing 1 wt% BHT (as an antioxidant) showed
 184 80% maintenance of the initial value over 6 months storage. The scintillator stability was improved
 185 with both BHT and post-curing treatment for 6 h at 58 °C.

186 The measured attenuation length of the post-cured BHT-containing plastic scintillator was
 187 approximately 0.9 m. The long-term stability of the attenuation length requires improvement, which
 188 will be attempted in a future study.

189 To increase the light yield, attenuation length, and long-term stability of these properties, we plan
 190 to develop as follows. Scintillators will be cured and post-processed with air interception. Others will
 191 use oxygen-purged resin material, and various types and contents of antioxidant agents will be tested.
 192 These modified new plastic scintillators will be evaluated and compared, with the purpose of realizing
 193 low-cost and stable materials.

194 Conflict of interest

195 Declarations of interest: none.

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 199 26800148, 17K18161, and 17K18316].

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- 279

280 281 **Figure Captions**

282
283 **Fig. 1.** Experimental setup of the light yield and attenuation length measurement. For the attenuation

284 length measurement, components that are fixed to motorized stage are moved every 1 cm. The

285 scintillators and the PMT surface are both covered with black sheets to shield them from ambient light

286 or from light emitted by either scintillator.

287 **Fig. 2.** Plastic scintillator samples measuring 40 mm × 75 mm × 3 mm. From left: BC408-1, NGT5-1,

288 and NGT6-1.

289 **Fig. 3.** Pulse shapes of the sample plastic scintillators observed by oscilloscope: (a) BC408-1, (b)

290 NGT1-1, (c) NGT2-1, (d) NGT3-1, (e) NGT4-1, (f) NGT5-1, and (g) NGT6-1. The irradiation

291 position of the β -ray was located 6 cm from the PMT surface on the centerline in the length direction

292 of the scintillator.

293 **Fig. 4.** ADC histograms of PH distribution of (a) BC408-1, (b) NGT1-1, (c) NGT2-1, (d) NGT3-1, (e)

294 NGT4-1, (f) NGT5-1, and (g) NGT6-1 irradiated by β -rays. The horizontal axis of (a) uses a scale

295 twice as large as that in the other figures.

296 **Fig. 5.** Long-term stability of the sample scintillators: (a) BC408, (b) NGT1, (c) NGT2, (d) NGT3, (e)

297 NGT4, (f) NGT5, and (g) NGT6.

298 **Fig. 6.** Attenuation length results (^{90}Sr source distance from PMT surface versus mean value). (a)

299 BC408L (day 0), (b) BC408L (day 365), (c) NGT6L-1 (day 33), (d) NGT6L-1 (day 280), (e) NGT6L-

300 2 (day 33), (f) NGT6L-2 (day 280).

301 **Fig. 7.** Long-term attenuation length stability.

302 303 **Table Captions**

304
305 **Table 1.** Plastic scintillator samples for light yield testing.

306 **Table 2.** Light yields and thicknesses of new plastic scintillators and BC408. The irradiation position

307 of the β -ray was located 6 cm from the PMT surface on the centerline in the length direction of the

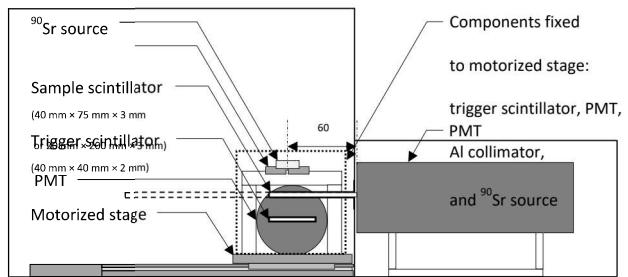
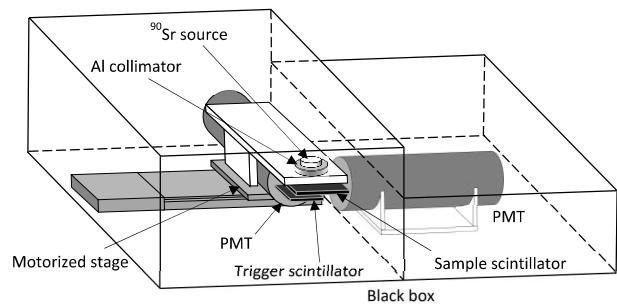
308 scintillator.

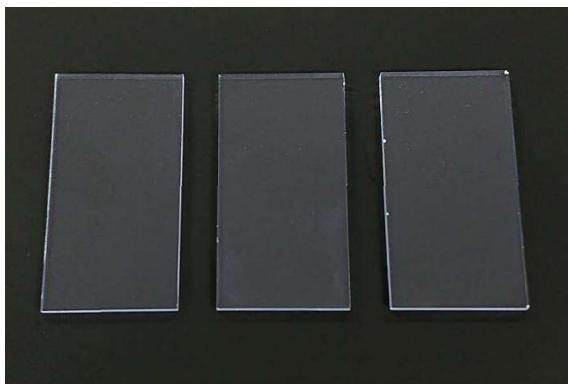
309 **Table 3.** Thickness of the sample scintillators, where Position x is the distance from the PMT surface

310 and Av represents the average thickness.

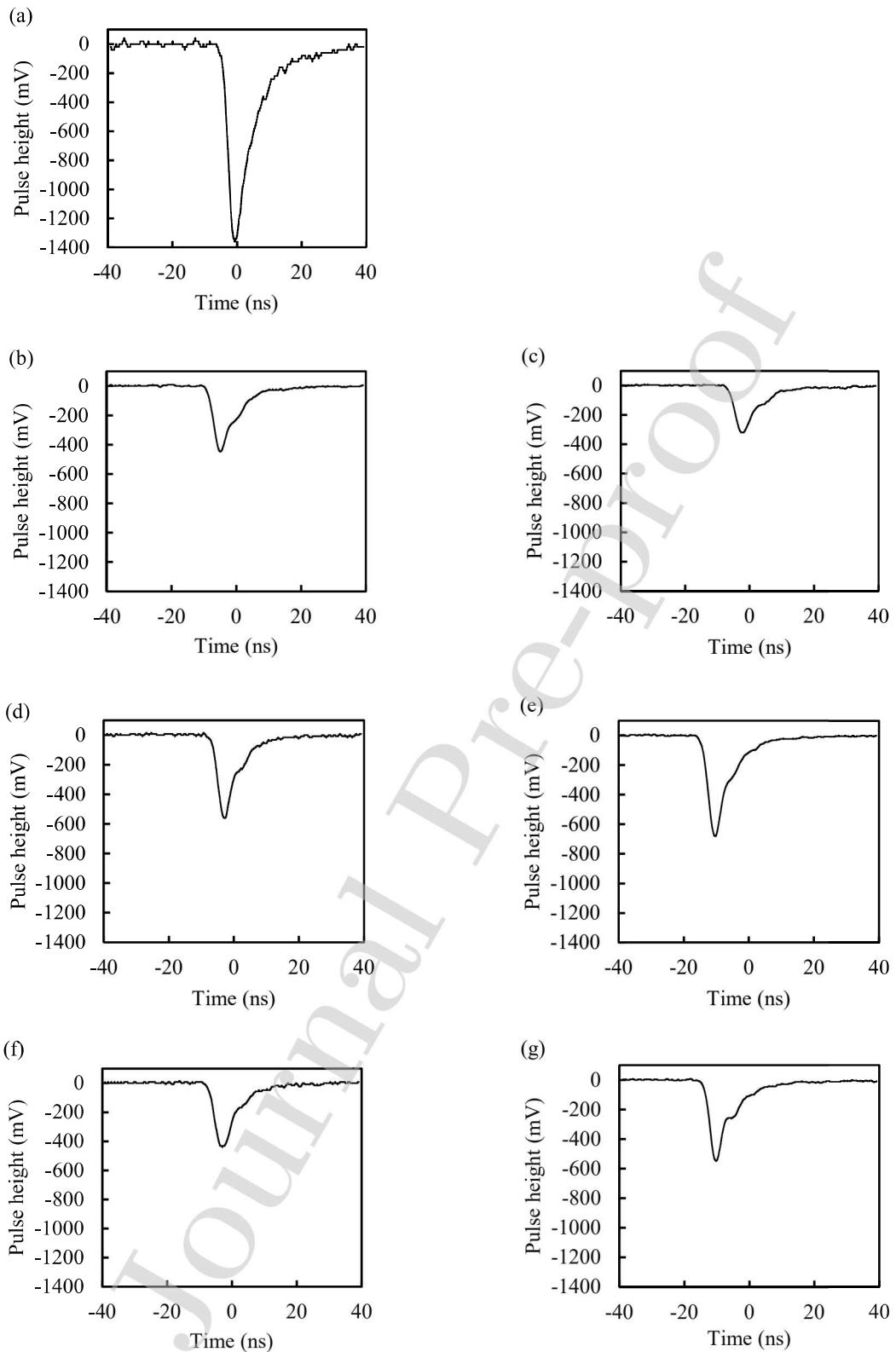
311 **Table 4.** Storage times of sample scintillators before measurement and attenuation lengths obtained

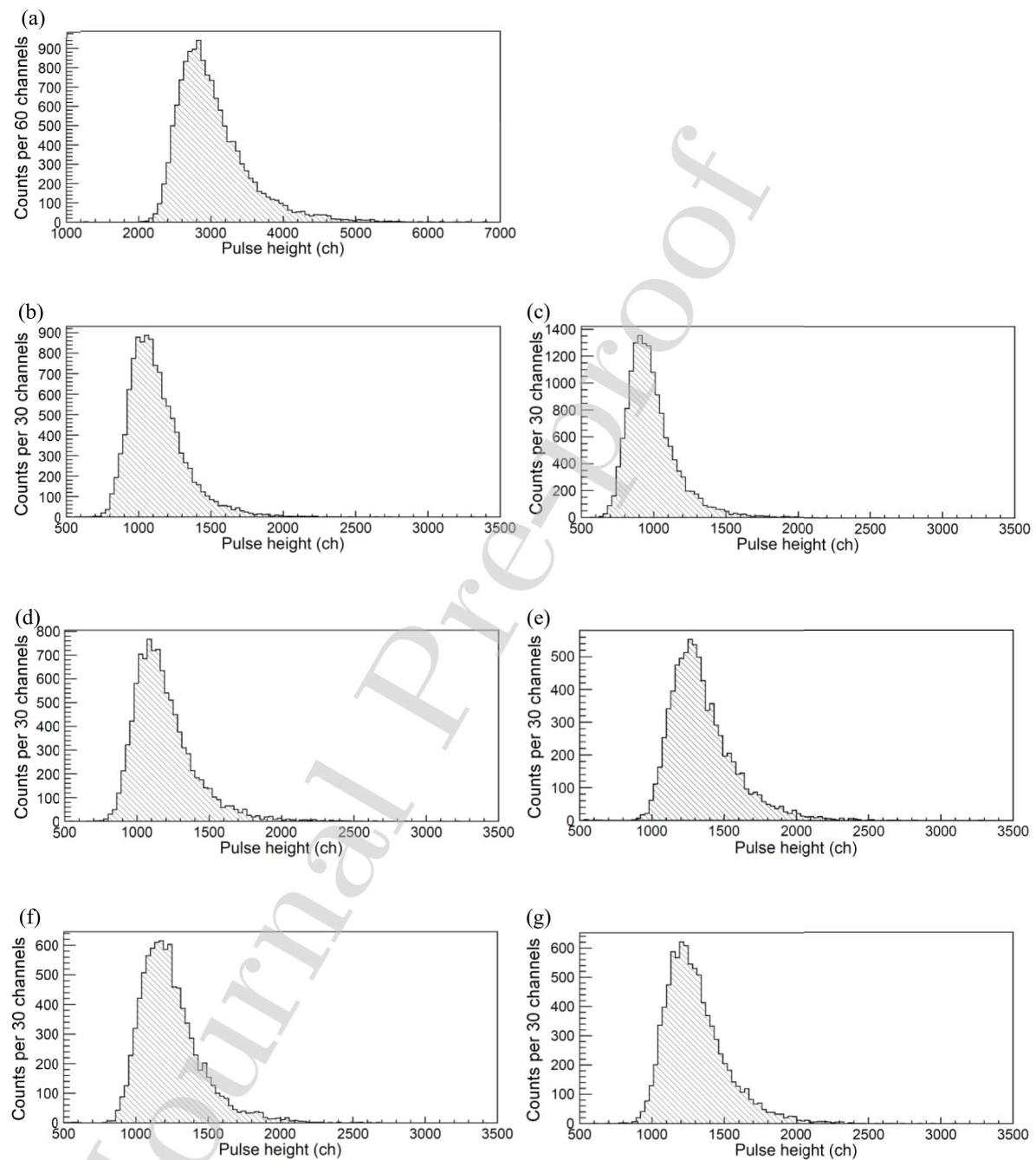
312 by function fitting. For BC408L, days elapsed since the first measurement are given. For NGT6L-1,2,
313 days elapsed since scintillator demolding are given.
314



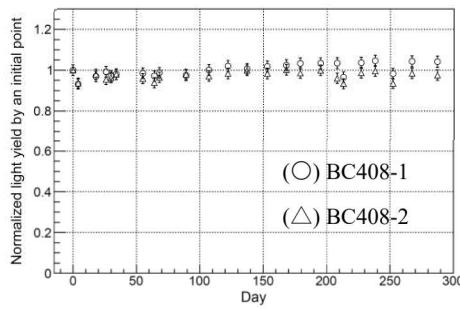


Journal Pre-proof

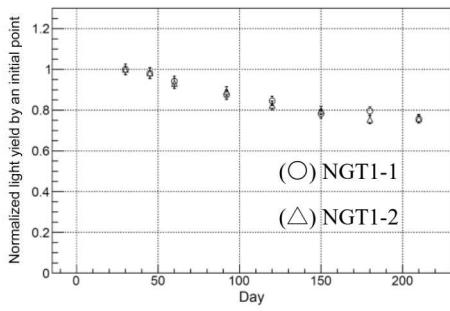




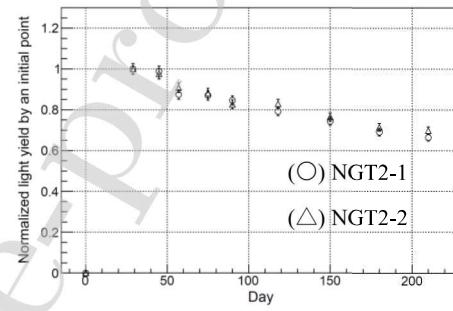
(a)



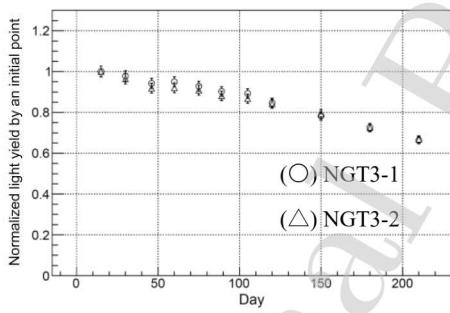
(b)



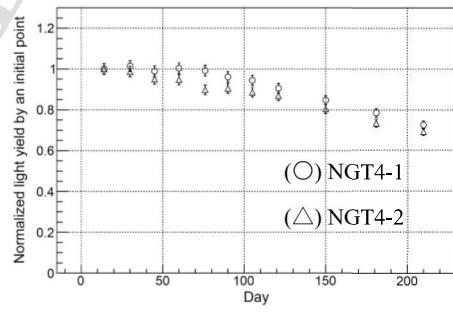
(c)



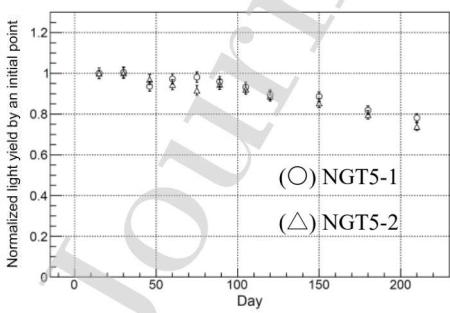
(d)



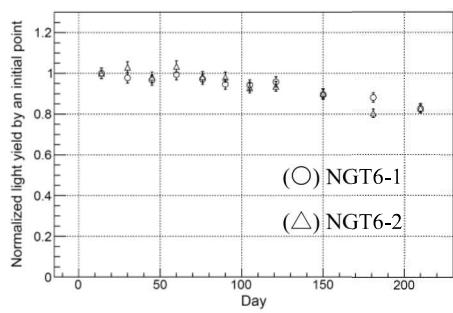
(e)

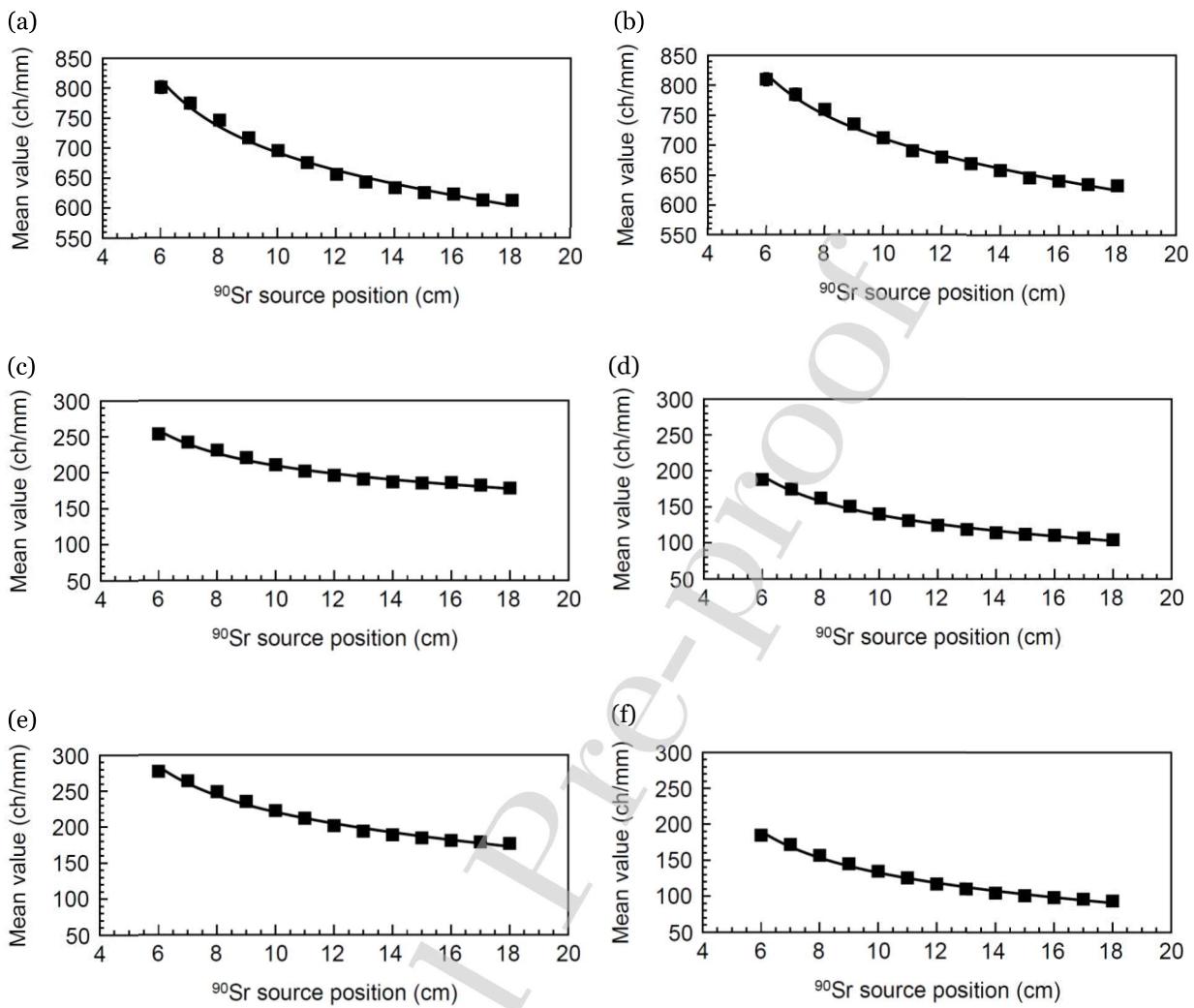


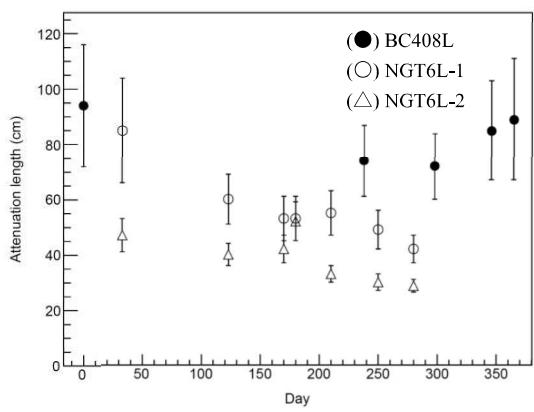
(f)



(g)







Sample name	BHT (wt%)	Post heating time (h)
NGT1-1, 2	0	0
NGT2-1, 2	0	6
NGT3-1, 2	0.1	0
NGT4-1, 2	0.1	6
NGT5-1, 2	1	0
NGT6-1, 2	1	6

Sample name		Normalized mean (ch/mm)	Mean value relative to BC408 (%)	Sample thickness (mm)
BC408	1	986 ±26	100 ±3	3.073
	2	988 ±26	100 ±3	3.116
NGT1	1	379 ±10	38 ±1	2.972
	2	372 ±10	38 ±1	2.955
NGT2	1	332 ± 9	34 ±1	2.973
	2	321 ± 8	33 ±1	3.052
NGT3	1	418 ±11	42 ±1	2.845
	2	452 ±12	46 ±1	2.977
NGT4	1	433 ±11	44 ±1	3.115
	2	417 ±11	42 ±1	3.094
NGT5	1	414 ±11	42 ±1	2.998
	2	396 ±10	40 ±1	3.093
NGT6	1	421 ±11	41 ±1	3.101
	2	405 ±11	42 ±1	2.990

Position x (cm)	BC408L (mm)	NGT6L-1 (mm)	NGT6L-2 (mm)
6	2.990	2.942	2.945
7	2.998	2.930	2.934
8	3.010	2.921	2.929
9	3.024	2.924	2.929
10	3.040	2.932	2.930
11	3.054	2.931	2.931
12	3.068	2.930	2.932
13	3.080	2.938	2.925
14	3.089	2.941	2.925
15	3.095	2.941	2.931
16	3.103	2.941	2.947
17	3.110	2.944	2.949
18	3.112	2.950	2.960
Av.	3.059	2.936	2.936

Sample name	Date of first measurement (days)	Date of last measurement (days)	Attenuation length of first measurement (cm)	Attenuation length of last measurement (cm)
BC408L	0	365	94 ± 22	89 ± 19
NGT6L-1	33	280	85 ± 19	42 ± 5
NGT6L-2	33	280	47 ± 6	29 ± 2